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Condensation and Storage of Hydrogen Cluster Ions

November 1988

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FOREWORD

This final report was submitted by the University of Dayton in support of contract F04611-83-C-0046 with the Air Force Astronautics Laboratory (AFAL), Edwards Air Force Base, CA. AFAL Project Manager was Lt Monte Turner.

This report has been reviewed and is approved for distribution in accordance with the the distribution statement on the cover and on the DD Form 1473.

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This report reviews the cluster ion approach to containerless condensation and introduces the concept of pseudo wall. An experimental pseudo wall device, designed for the study of containerless condensation techniques, is described. Calculations indicate that the proposed device will be capable of confining cluster ions in the mass range 2-10^7 for time exceeding 10^4 seconds. A "next generation" device is projected to be capable of containing a total mass of 0.3 milligrams.

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SUMMARY

This report briefly reviews our current understanding of the Cluster Ion (CI) approach, describes an experimental apparatus that will (for the first time) allow long-term studies ($>10^4$ seconds) of very large hydrogen cluster ions ($1<m/q >10^{10}$) in a very high density nonneutral plasma (densities exceeding the Classical Brillouin Limit), and makes recommendations for future work.

The central problem is the "containerless" and "collisionless" assembly of subatomic, atomic, and molecular fragments into bulk antimatter, in this case solid antihydrogen "ice." It is necessary to condense antimatter into this form to achieve favorable payload fractions (when it is used as a fuel for propulsion) and to have a new means of high-density energy storage (when it is used for energy storage). Since this condensation has not been done before and must be done under "containerless" and "collisionless" conditions (with regard to collisions with ordinary matter), severe restrictions are placed on the methods used. Fortunately, <u>normal</u> matter can be used as a simulant in all aspects of the investigation, since the only relevant difference in the two forms of matter in this case is the sign of the atomic charge (ignoring parity and time). All work described in this report will refer to <u>normal</u> hydrogen, unless otherwise denoted.

As a result of our research for the past two years, an appropriate condensation method is now known whereby high density clusters may be grown via "containerless" condensation methods. This method, the CI approach, uses condensation of suitable reactants upon cold, trapped, hydrogen cluster ions.

To perform this "containerless" condensation (CC), cold fragments such as hydrogen atoms, molecules, and ions must be formed (resulting in the release of significant amounts of chemical energy) that have little or no translational, vibrational, or rotational energy. These cold fragments are then assembled to form ever larger fragments. The end result of this research will be a detailed and experimentally established "containerless" synthetic method.

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LIST OF SYMBOLS

2α	# nuclei in a cluster ion or mass of a cluster per proton mass
α	# molecules in a cluster ion
n	number density cm^{-3}
V	volume
v	velocity
N	# of cluster ions
M	mass, critical size for M^+ process
a	radius of sector magnet, S.C. magnet radius, critical site A^+, central conductor radius
D	cm^3/molecule for hydrogen cluster ions
z	charge per cluster, axial distance from center of S.C. magnet
Z	distance, spacing between electrodes in Wien filter
L^o	mean free path
V'	potential
β	charge state $\pm$
f	collision frequency
L'	attenuation coefficient
e	charge of electron
c	conductances (L/s)
I	current (amps)
J	flux
A	area, *Einstein* A
B	magnetic field
E	electric field, energy
ϕ	potential on central rod

Ω, w_c	cyclotron frequency
d	radius of trap
$R^{\pm}$	annular plasma inside (R^-) and outside (R^+) radii
k	Boltzmann constant, integer
T	temperature
Γ	plasma state parameter
r_{ci}	cluster ion radius
r_L	Larmor radius
q	charge
S_p	speed of pump
P_s	source pressure
P_t	trap pressure
α_w, R_w	outgas rate (watts/m^2)
A_w	wall area
w	slit width
R	radius of curvature
L	length of S.C. magnet (axial)
Q	cross section, throughput of pumping section
r	radius of annular plasma
p	integer
Δ	thickness of plasma (annular)
w_p	plasma frequency
v_θ^o	azimuthal velocity
E_r^o	radial electric field
u	mass to change ratio

INTRODUCTION

The future development of high-power, high-flux accelerators is projected to result in the eventual production of significant annual yields (milligrams) of antiprotons and positrons. The availability of this new form of matter suggests a fascinating new science having many applications. One application of this new substance (in the area of energy storage) uses it as a propellant with unsurpassed characteristics (payload fractions, specific impulse, thrust etc.) which make it ideally suited for missions currently impossible. However, before it can be used in this application, it must be put into a high-density form; it must somehow be nucleated (or condensed) into a form that leads to high energy density and is easily confined (levitated). Fig. 1 is a diagram of the approach for the past two years. R. L. Forward (Refs. 1,2) has suggested frozen parahydrogen (levitated) as an ideal form; however the means by which one obtains this product (starting with trapped protons and electrons) was unknown. Hence, project research for the past two years sought a means of bridging the enormous gap between individual, unrecombined fermions and bulk matter. The CI approach to "Containerless Condensation" (CC) soon emerged as a feasible approach within constraints imposed by present-day and near-term technology. The CI approach uses the growth and manipulation of hydrogen cluster ions in traps using electric and magnetic fields. The main advantages of cluster ions are these:

1. They are more stable than uncharged clusters (except H_2^+ versus H_2). Cluster ions with mass to charge ratios beyond $u=10^5$, have been observed experimentally (Refs. 3,4).

2. They can be trapped, transported, and stored in devices that use the Coulomb force (trap times of months and depths greater than 10 eV are typical) (Refs. 5,6).

3. They have very large rate constants for collision with a neutral antiparticle ($>10^{-9}$ cm^3 s^{-1} $particle^{-1}$).

4. They have a dipole-allowed IR spectrum and thus may radiatively cocl their vibrational degrees of freedom (except H_2)(Einstein A's ~ 10^3

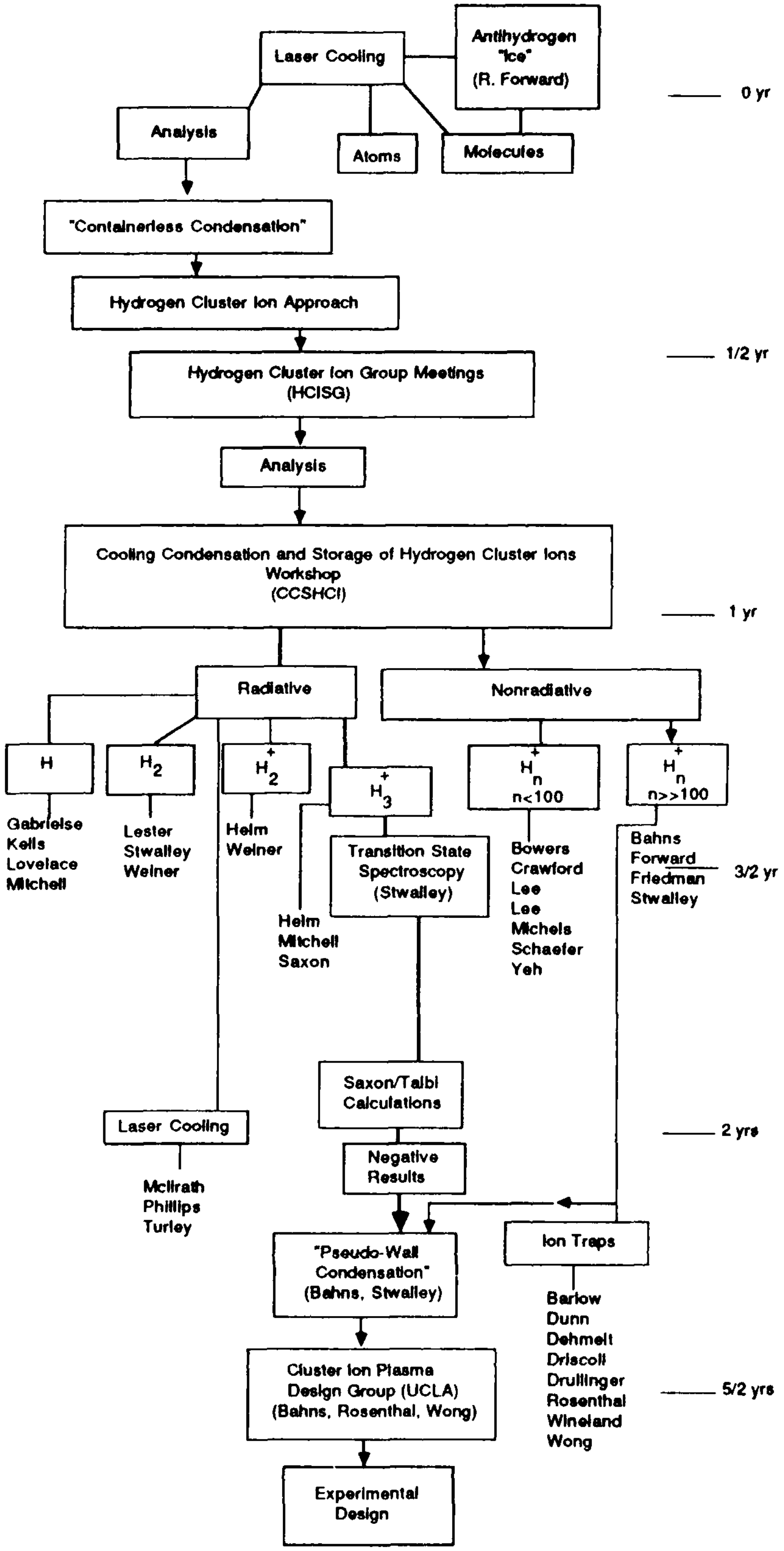


Figure 1. Flowchart of Events that Led to the Proposed Plan of Research. Also given are the names associated with each category.

s^{-1}). The spectroscopy of normal H_3^+ (the analog of H_3) has been reported (Refs. 7-10). The radiative properties have been reviewed, (Refs. 11-13) as have large cluster ions (Ref. 14).

5. Their translational degrees of freedom may be cooled by existing (nonlaser) techniques.

The HCISG (Hydrogen Cluster Ion Study Group) meetings (Ref. 15) reinforced enough of the original notions to deserve a workshop (called CCSHCI, for Cooling Condensation and Storage of Hydrogen Cluster Ions) devoted to a more in-depth look. Three general strategies emerged: radiative, nonradiative, and both. For the most part, there seem to be no promising stimulated radiative channels (and few spontaneous channels), due primarily to the general lack of bound excited electronic states in these systems, as shown in recent calculations on H_3^+ (Ref. 16). Hence, one is led to consider nonradiative pathways with the hope that spontaneous pathways will take care of themselves. The pseudo-wall concept devised by the author and Professor W. C. Stwalley has been implemented (in collaboration with Professors A. Y. Wong and G. Rosenthal) as discussed in the Experimental Design section.

The Neutral Reactants: H and H_2

A brief review of these molecules in the context of CC is presented. How to proceed with the radiative formation of the simplest antifragments (H, H_2^+, H^-, H_2) is still rather uncertain. For H_2^+ formation see Helm's report (Ref. 17). Since H^- does not have discrete excited states, no way is known to manipulate it radiatively. The formation of H^- may work well in a pseudo-wall environment. The small negative cluster ions have been removed from consideration due primarily to their instability (Refs. 13, 18).

Hydrogen atoms can be formed via stimulated recombination (the most direct path being continuum-2p) followed by spontaneous radiation (2p-1s) to the ground state (Fig. 2) (Refs. 19, 20). Hydrogen atoms may also be laser cooled to near the quantum limit (Ref. 21), and otherwise laser

size were available, one might use them as a substrate for the recombination of hydrogen atoms. Ideally, one would like to nucleate hydrogen atoms directly onto (or into) a cluster ion. Research is needed that reveals how the recombination energy is coupled into the cluster ion. Once formed, however, the means of trapping the atoms are known (Refs. 23-25).

Unless a breakthrough occurs, any nucleation process that uses hydrogen molecules is likely to be complicated and inefficient. A specific nucleation pathway that uses H and avoids H_2 is actively being sought. For H_2 to be useful in the nucleation of the smallest hydrogen cluster ions (say, $u<29$), it must be quite cold in all of its degrees of freedom, which is difficult to accomplish. Further difficulties are that H_2 cannot radiatively cool itself in its ground state (without the help of a third body) and we know of no simple way of laser cooling any of its degrees of freedom. H_2 formation is difficult and inefficient and once formed there is no known way of keeping it from annihilating on the walls (see report by K. M. Sando, in CCSHCI proceedings). The means of making it radiatively are currently being studied (Ref. 26). For a discussion of the intramolecular potential, see Lester (Ref. 27).

CLUSTER ION APPROACH TO CONTAINERLESS CONDENSATION

This section is devoted to topics that relate in one way or another to the development of CC techniques.

Hydrogen Cluster Ions: Review of Structure and Properties

As stated in the Introduction, the properties and structure are well known for the smallest cluster ions and have been reviewed extensively (Stwalley (Ref. 28) has prepared an extensive bibliography; see Appendix B). Some relevant properties of cluster ions are included in Table 1 (Ref. 29).

As a review, we are considering just the positive cluster ions. This was decided because the small negative cluster ions (excluding H^-) are unstable to spontaneous detachment. Although it is known that negative hydrogen cluster ions become stable at some (unknown) critical size, they would still be sensitive to photodetachment, making them less desirable (Ref. 13).

In current penning-type trapping systems a negative ion plasma is more difficult to deal with. With a positive ion plasma, a small electron population will simply move to the ends of the trap and be removed by the end termination electrodes. With a negative ion plasma, a small population of electrons would not be able to leave the system. These trapped electrons add charge to the system and introduce the possibility of two species diochotron instability. Such instability would probably reduce confinement time.

The small singly charged, positive hydrogen cluster ions are well studied both experimentally and theoretically, and are all known to be stable. The doubly charged hydrogen cluster ions are stable for sizes larger than about 2000 (this may be inferred from data taken from Yano [Ref. 4]). Calculations predict the dipositive ions to be stable at size 887, tripositive ions at 2906, and the tetrapositive ions at 6196

TABLE 1. Overview of Particle (and Antiparticle) Properties and Characteristics. (From Stwalley [Ref. 29]).

particle	e^-	H^+(:p)	H	H^-	H_2^+	H_2	H_3^+	H_3	H_4^+	H_4	$H_{N≠}^+$	$H_{N≠}$
antiparticle	e^-	$\bar{H}^+(\bar{p})$	$\bar{H}$	$\bar{H}^-$	$\bar{H}_2^+$	$\bar{H}_2$	$\bar{H}_3^+$	$\bar{H}_3$	$\bar{H}_4^+$	$\bar{H}_4$	$\bar{H}_{N≠}^+$	$\bar{H}_{N≠}$
binding energy in eV	-	-	13.6	0.75	2.6	4.5	4.1	0	0.1	<0.01	...0.008	0.008
products	-	-	H^+,e^-	H,e^-	H^+,H	H,H	H^+,H_2	H,H_2	H,H_3^+	H_2,H_2	H_{N-2}^+,H_2	H_{N-2},H_2
sensitivities†:												
λ < 0.2 μ VUV	-	-	x	x	x	x	x	x	x	x	x	x
0.2-0.4 μ UV-VIS	-	-	-	x	x	-	(x)	-	(x)	-	x	x
0.7-100 μ IR	-	-	-	-	-	-	-	-	-	-	x	x
λ > 100 μ microwave	-	-	-	-	-	-	-	-		-	x	x
radiative recomb. (e^-)	*	x	x	-	(x)	-	(x)	-	(x)	-	(x)	
dissoc. recomb. (e^-)	*	-	-	-	x	-	x	-	x	-	x	-
assoc. detach. (H^-)	-	-	x	*	-	-	-	x	-	-	-	-
rxn. (H)	-	-	*	-	x	-	x	(x)	x	-	-	-
rxn. (H_2)	-	-	-	-	x	*	-	-	x	-	-	
neutralization (H^-)	-	x	-	*	x	-	x	-	x	-	x	
radiative assoc. H, H_2)	-	-	-	-	-	-		-	-	-	x	x
trapping:												
Penning/Paul	x	x	-	x	x	-	x	-	x	-	x	
laser	-	-	x	(x?)	x?	x?	-	-	-	-	-	-
inhomogeneous magnet	-	-	x	-	-	-	-	-	-	-	x	x
cooling:												
radiative	-	-	-	-	-	-	x	-	x	-	x	x
laser	-	-	x	-	x?	x?	?	-	?	-	-	-
lepton (e^-)	x	x	-	x	-	-	-	-	-	-	?	-
transfer:												
electrostatic	x	x	-	x	x	-	x	-	x	-	x	-
magnetic	-	-	x	-	-	-	-	-	-	-	-	x
laser	-	-	x	-	(x?)	(x?)	-	-	-	-	-	-

≠ N assumed large (>10^3).

† All collision partners assumed to be cold.

* Reactant.

Refs. 30, 31). The odd-numbered cluster ions consist of an H_3^+ core, surrounded by H_2 molecules. The even-numbered cluster ions are similarly structured but have one attached H atom.

Large Hydrogen Cluster Ion Sizes and Trapping

Taking the density of solid hydrogen (0.076 g cm^{-3}) to represent the density of a large cluster ion, one obtains the constant D = 5 x 10^{-23}

cm^3/molecule, which was used to construct Table 2. In principle, if the correct source of hydrogen cluster ions is developed, virtually any sized cluster ion may be trapped in the proposed ion trap, up to roughly

TABLE 2. Large Hydrogen Cluster Ion Diameters.

Cluster # (# of molecules)	Log (cluster diameter) (microns)
10^4	-2
10^7	-1
10^{10}	0
10^{13}	1
10^{16}	2
10^{19}	3

10^{14} atoms. This is discussed further in the section entitled "Very Cold Hydrogen Cluster Ions."

Containerless Condensation by Pseudo-Wall Reactions

The ideal way to perform containerless condensation is to arrange conditions whereby both radiative and nonradiative processes occur. This section briefly discusses the concept of a pseudo-wall and presents a number of condensation pathways based on this concept.

Introduction to Pseudo-Walls

The primary goal is a detailed understanding of the available association channels of hydrogen cluster ions. The simultaneous requirements of the association reactions are the release of energy,

conservation of momentum, and generation of a larger cluster ion. Generally, the association pathways may be categorized as radiative, for example,

$$H_2^+ + H_2 = H_4^+ + hv \tag{1a}$$

or nonradiative,

$$H_2^+ + H_2 = H_3^+ + H \tag{1b}$$

Both of these reactions are exothermic. However, in reaction (1a), a larger cluster ion is obtained because the channel allows the excess energy (kinetic, electronic, etc.) to be radiated as a photon. In case (1b), the excess energy is released in the form of kinetic energy of the ejected atom. Certain channels may exist that involve both kinds of processes simultaneously. These channels are still being explored.

One way of ensuring that collisional and radiative relaxation channels are favored is to manipulate the reactant densities and temperatures, i.e., have a reaction that always involves one (or both) of the reactants in a special condensed phase, referred to as a "pseudo-wall." A pseudo-wall is a cold, trapped, high-density phase that is thermally connected (via E and M fields) to the environment. This cold, high-density condensed phase results in collective effects that may occur on short time scales; it behaves somewhat like a wall both kinetically and thermodynamically. This concept is really just an extension of the liquid and solid pure electron plasmas that have been talked about for some time (Refs. 32, 33). The relevant parameter is

$$\Gamma = e^2/(a\ kT),$$

where a is given by $[(\frac{4}{3})\ \Pi\ a^3\ n] = 1$, e is the charge, T is the temperature, n is the density, and k is Boltzmann's constant. Γ=1 defines a plasma, at Γ=2 one has a liquid, and finally at Γ=155 one obtains a solid pure electron plasma. Γ is a measure of the Coulomb potential to the

thermal kinetic energy in the plasma. One wants the largest range of Γ obtainable. This may be done by varying T and n.

For the proposed device, the achievable density will be roughly $n=10^8$ cm^{-3} for clusters smaller than 10^4 atoms. If the T=4 K, one obtains

$$\Gamma \sim 120,$$

which falls well within the liquid regime.

When a single incoming (neutral or charged) particle collides with the pseudo-wall, part of the collision energy is absorbed, almost instantaneously, by neighboring particles that make up the wall. The other important feature is that, since the pseudo-wall is connected to an ion trap by E & M forces, it can transfer its energy to the environment. When referring to the above reactions, if just H_2^+ is made into a pseudo-wall, they become

$$\{H_2^+\} + H_2 = H_4^+, \tag{2a}$$

and

$$\{H_2^+\} + H_2 = H_3^+ + H. \tag{2b}$$

Here, we consider the reaction of H_2 molecules within the cold, high density environment of $\{H_2^+\}$ (T<4 K and $n=10^{11} cm^{-3}$). The importance of this process is that the collisions are not strictly 2-body, but may be 3-body (and higher, up to 10^{14} body for the proposed pseudo-wall). This results in substantial collisional energy transfer (rather than just radiative) of the energy of the exothermic reaction to the pseudo-wall (and ultimately to the environment). The advantage illustrated in this example is that reaction (1a) cannot proceed radiatively, while (2a) can. In this manner, a finite set of reaction pathways result that may proceed much more efficiently and at much higher rates (coming from the inclusion of collisional relaxation mechanisms, and high effective densities).

The conditions that specify a pseudo-wall are now defined. The main constraint is (L<<r), where L^o is the collisional mean free path of the cluster ion and a (assume a=1 cm) is the physical radius of the pseudo-wall. If $L^o=.01a=10^{-2}$ cm, and the collisional cross-sections (Q) are at least 10^{14} cm^2, the pseudo-wall density (n) is obtained from

$$L^o = .001a = 1/(nQ),$$

giving a neutral density $n=10^{16}$ cm^{-3} (the temperature is adjusted to achieve this density). In contrast, the maximum densities of ions in non-neutral plasmas are 10^{11} cm^{-3}. Under these conditions a single cluster ion in a pseudo-wall would undergo roughly 10^6 bimolecular collisions per second with neutral particles. Constructing such a cylindrically shaped pseudo-wall of volume (V=30 cm3), would require only about N=nV, or 3 x 10^{12} cluster ions.

Another way of specifying pseudo-wall conditions is to require that the collision frequency, f, be greater than the radiative lifetime, t, of any excited states, ensuring collisional deactivation at some velocity v: $f=nQv > 1/t$. This criterion is satisfied for neutral densities, n, of 10^{16} cm^{-3}, cross sections, Q, of 10^{-14} cm^2, and velocities of 10^4 cm s^{-1} for radiative lifetimes of 10^{-6} s and longer (cluster ions have radiative lifetimes of several milliseconds, for vibrational transitions).

The maximum cooling power required of the pseudo-wall may be estimated. The most exothermic reaction is the first one in Fig. 3, requiring 13.6 eV for each H atom. Assuming a nucleation rate of 10^{12} particles per second (166 nanoamps), this results in 13.6 X 10^{12} eV s^{-1} (or 2 micro-watts) of recombination energy that must be coupled through the pseudo-wall to the environment. Established resistive cooling techniques will be used, if necessary.[34]

The main reactions which may take place in pseudo-walls are illustrated in the reaction matrix of Fig. 3. Certain working assumptions were made in the construction of this matrix:

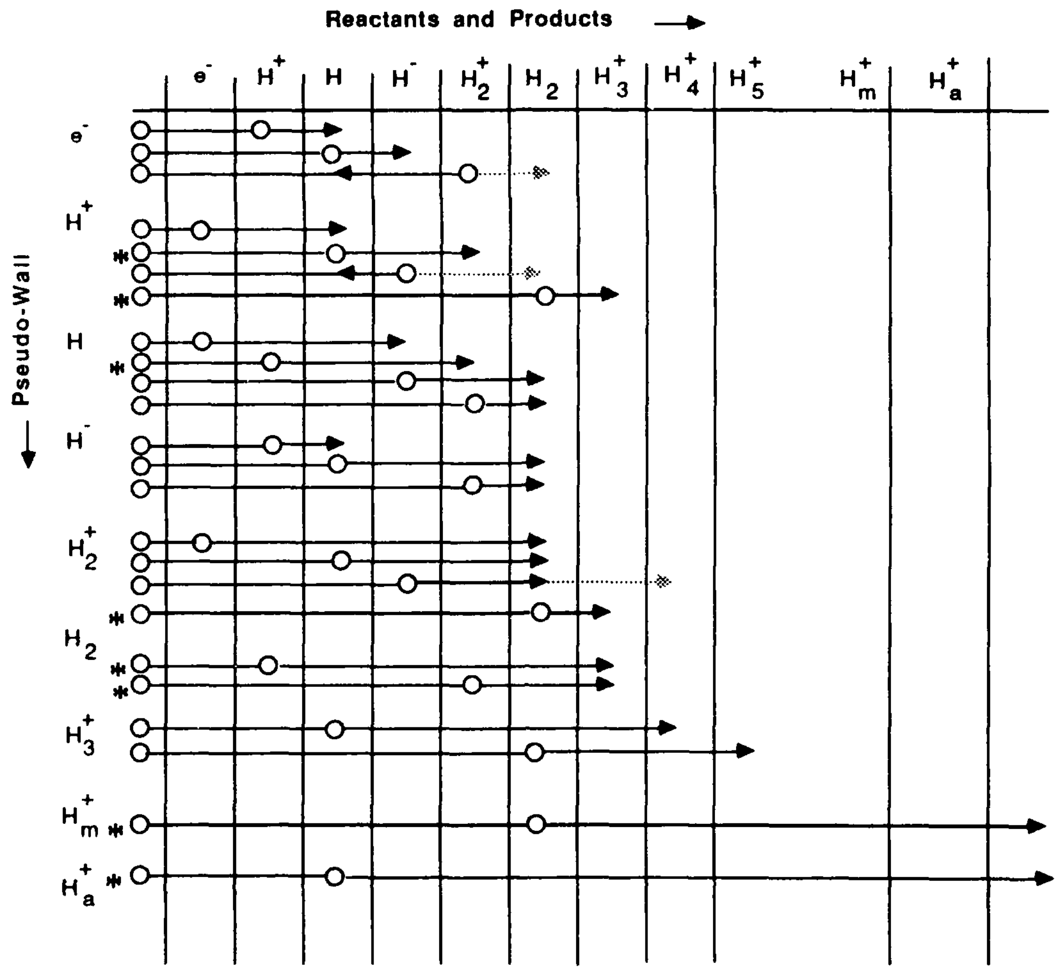


Figure 3. Reaction Matrix Showing Containerless Condensation Routes. In the first reaction, protons are added to a pseudo-wall composed of electrons, resulting in the production of cold hydrogen atoms.

1. All particles (one or both reactants) can be turned into pseudo-walls; ionic pseudo-walls are currently preferred.

2. Only two-reactant combinations are considered (reactant plus pseudo-wall).

3. No nuclei (other than deuterium) larger than the proton.

4. Minimum dependence on radiative processes (photons are excluded).

5. No anions other than H^-.

6. No obvious fragmentation reactions.

7. No unstable or short-lived particles (for example positronium, H_2^-, H_3^-, etc.).

Some reactions in Fig. 3 (with asterisk) could proceed spontaneously within the same pseudo-wall to grow large cluster ions. Recall that the cluster ions of sizes H_m^+ and H_a^+ are the threshold cluster ion sizes for ion-molecule (M^+) and ion atom (A^+) association, respectively. These have been postulated to exist at sizes around 29 and 2000 respectively.[15] The objective is to select the optimum pathway to these large cluster ions by connecting reactions and defining pseudo-walls.

Condensation Pathways Using Pseudo-Walls

Fig. 4 illustrates one of the many possible condensation pathways. One example (out of many) of a "containerless" condensation pathway is designated by the large arrows in Fig. 4. The proposed experimental approach is designed to verify the reactions and pathways of this matrix. The most fundamental reactions are those involving either H^+ or e^- which react to produce H, H^-, or H_2^+ (the production of H_2 may also occur in these cold plasmas but this is assumed minimal). The reactions indicated by the condensation pathway in Fig. 4 may be written specifically as

1. $(e^-) + H^+ = H$

2. $(e^-) + H = H^-$

3. $(H^-) + H = H_2 + e^-$

4. $\{H^+\} + H = H_2^+$, $\{H_2^+\} + H_2 = H_3^+ + H$, and

5. $\{H_3^+\} + H_2 = H_5^+$, etc.

The procedure is repeated until a critically sized cluster ion is obtained such that it will undergo repeated additions of H_2 without fragmentation. This may be written

$$H_m^+ + H_2 = \{H_{m+2}^+\}.$$

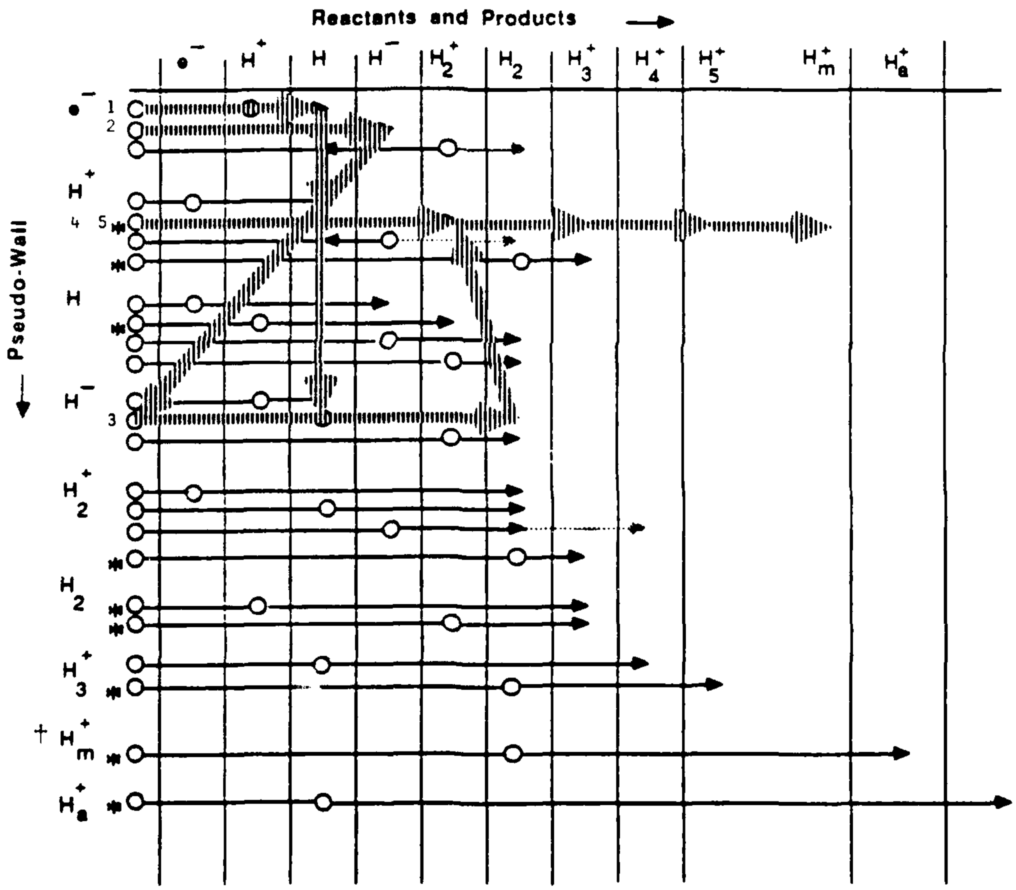


Figure 4. Reaction Matrix with a Potential CC Pathway. (Note large arrows.) The reaction denoted by (†) may be written, $H_m^+ + pH_2 = H_a^+$, where p is an integer. Refer to text for reactions denoted with numbers.

H_m^+ is referred to as the threshold seed cluster ion size for the m^+ process. If this process is repeated enough times another seed critical cluster ion size will be reached such that the reaction

$$(H_a^+) + H = (H_{a+1}^+),$$

will proceed. Experiments have been proposed that will determine the cluster ion sizes H_a^+ and H_m^+. A few other possible pathways have been illustrated in Figs. 5 to 7.

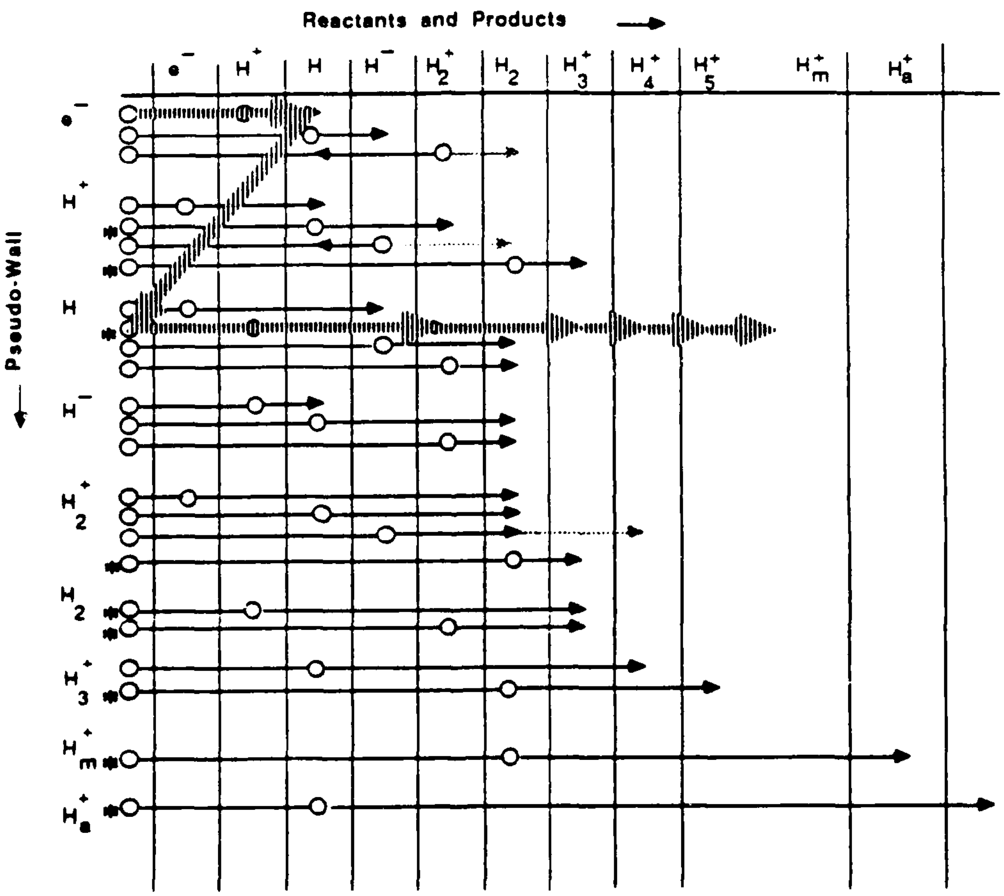


Figure 5. Reaction Matrix with a Potential CC Pathway. (Note large arrows.)

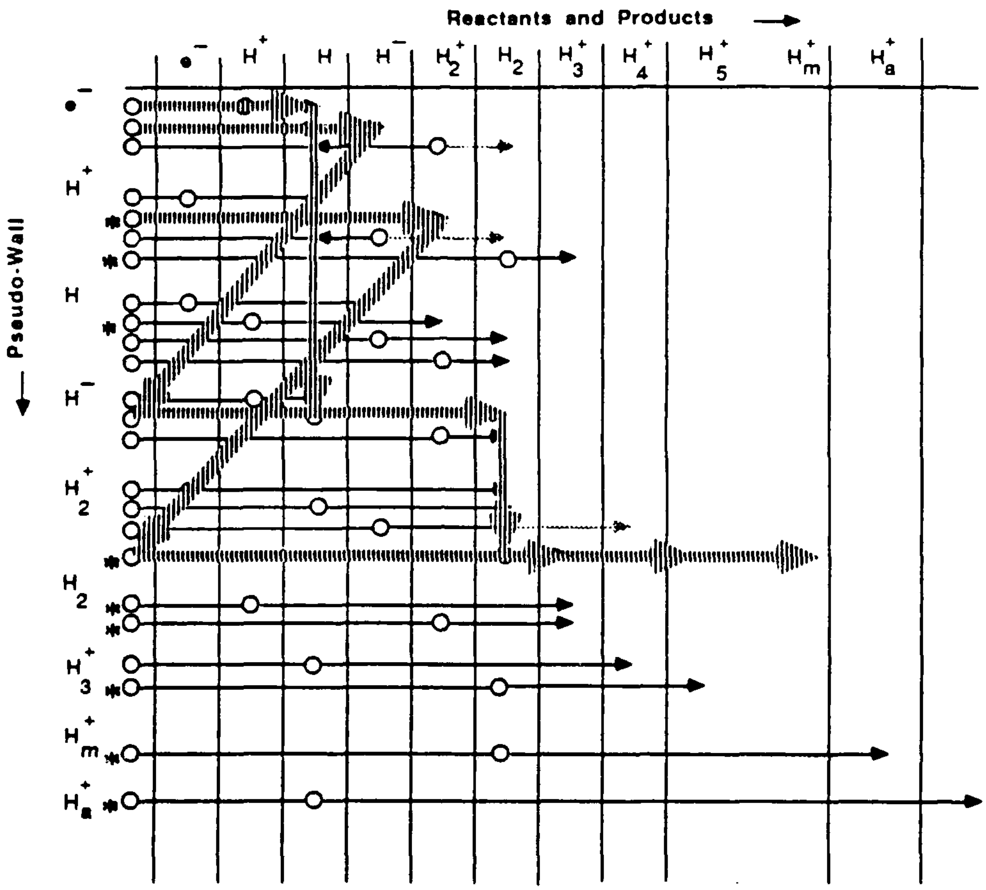


Figure 6. Reaction Matrix with a Potential CC Pathway. (Note large arrows.)

Destruction of Hydrogen Cluster Ions by Annihilation

This section discusses the various ways hydrogen cluster ions may be destroyed or removed from the trap due to collisions (resulting in surface annihilation) with neutral (and nonneutral) antihydrogenic species (made up of fermions, these represent a main constituent of background gas).

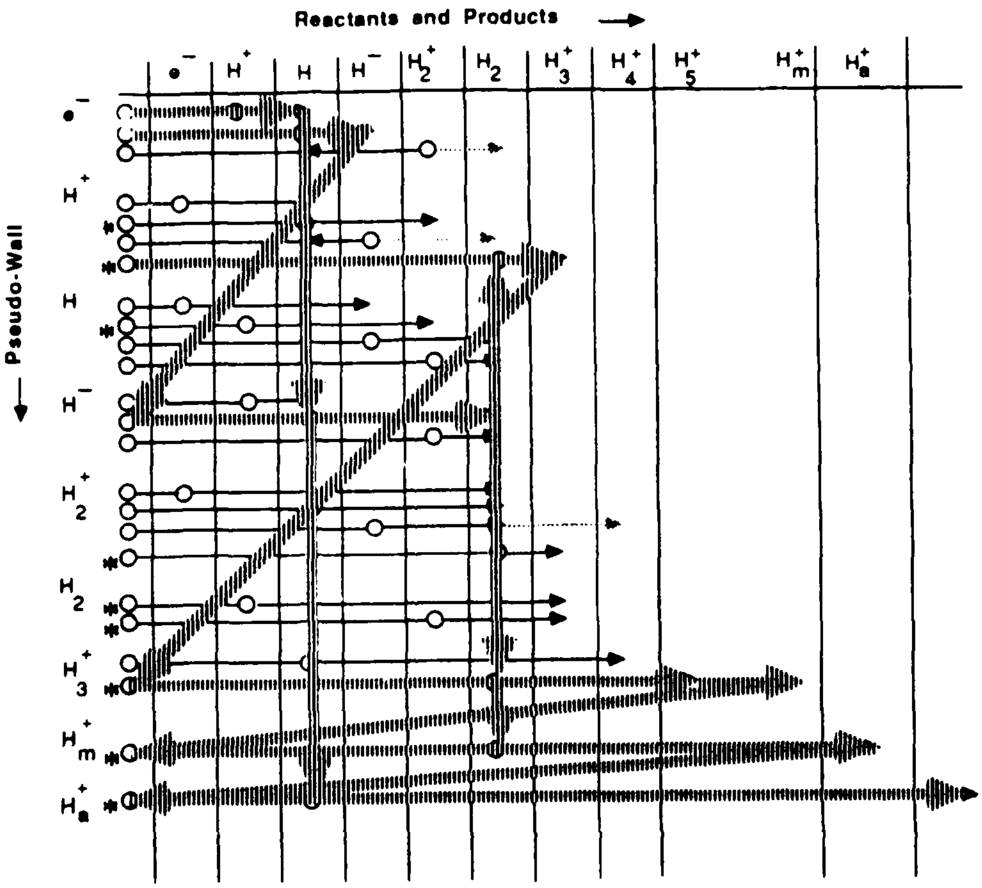


Figure 7. Reaction Matrix with a Potential CC Pathway.
(Note large arrows.)

Considering just the primary event, the only annihilation products to consider are high-energy pions (Refs. 1, 2) from the proton-antiproton annihilation and Γ rays from electron-positron annihilation. It is assumed that the coupling of motion between Pions and the molecular (and atomic) degrees of freedom is small, especially vibration, rotation, and translation (sources of pion-electron cross sections for electronic excitation, ionization, should be found). The 1/e attenuation coefficient for γ rays is much larger (L'(1/e) >128 cm, E>0.5 MeV) than the dimensions of the cluster ions or plasmas and may be neglected.

There are other ways of destroying cluster ions: recombination, photodissociation, etc.; these however have been considered elsewhere (Refs. 9, 35). The reactions to be considered here are

1. $\overline{H_n^+}$ + low energy e^- <= Coulomb Barrier,

2. $\overline{H_n^+}$ + H (or $\bar{H}_2$) => $\overline{H_{n-1}^+}$ (or $\overline{H_{n-2}^+}$)

+ annihilation products, and

3. $\overline{H_n^+}$ + P^+ => $\overline{H_{n-1}^0}$ + annihilation products, where the bar denotes antimatter (note that $\overline{H_n^+}$ is negatively charged).

Reaction 1 may be neglected for low-energy positrons.

Reaction 2 under the above assumptions will not yield fragments, but will only reduce the mass of the cluster ion by one unit; it too is neglected.

Reaction 3 is the most problematic since it leads to the production of a neutral cluster ion, H_{n-1}^0 (only slightly less massive). Here, we must consider the cross section for proton-antiproton annihilation. Considering destruction by just low energy (thermal T=300 K) antiprotons, an extrapolation of the data (Ref. 36) from Fig. 8 (upper right) gives a cross section of 10^9 barns or 10^{-15} cm^2. The density of protons in a cluster ion is roughly 10^{22} cm^{-3} so that the mean free path of an antiproton in a cluster ion can be estimated to be

$$L = 1/(10^{-15} \times 10^{22}) = 10^{-7} \text{ cm}.$$

This means one need only consider surface annihilation from thermal antiprotons since the cluster ions will, in general, be much larger than 1 nm. Since the populations and annihilation cross sections for higher

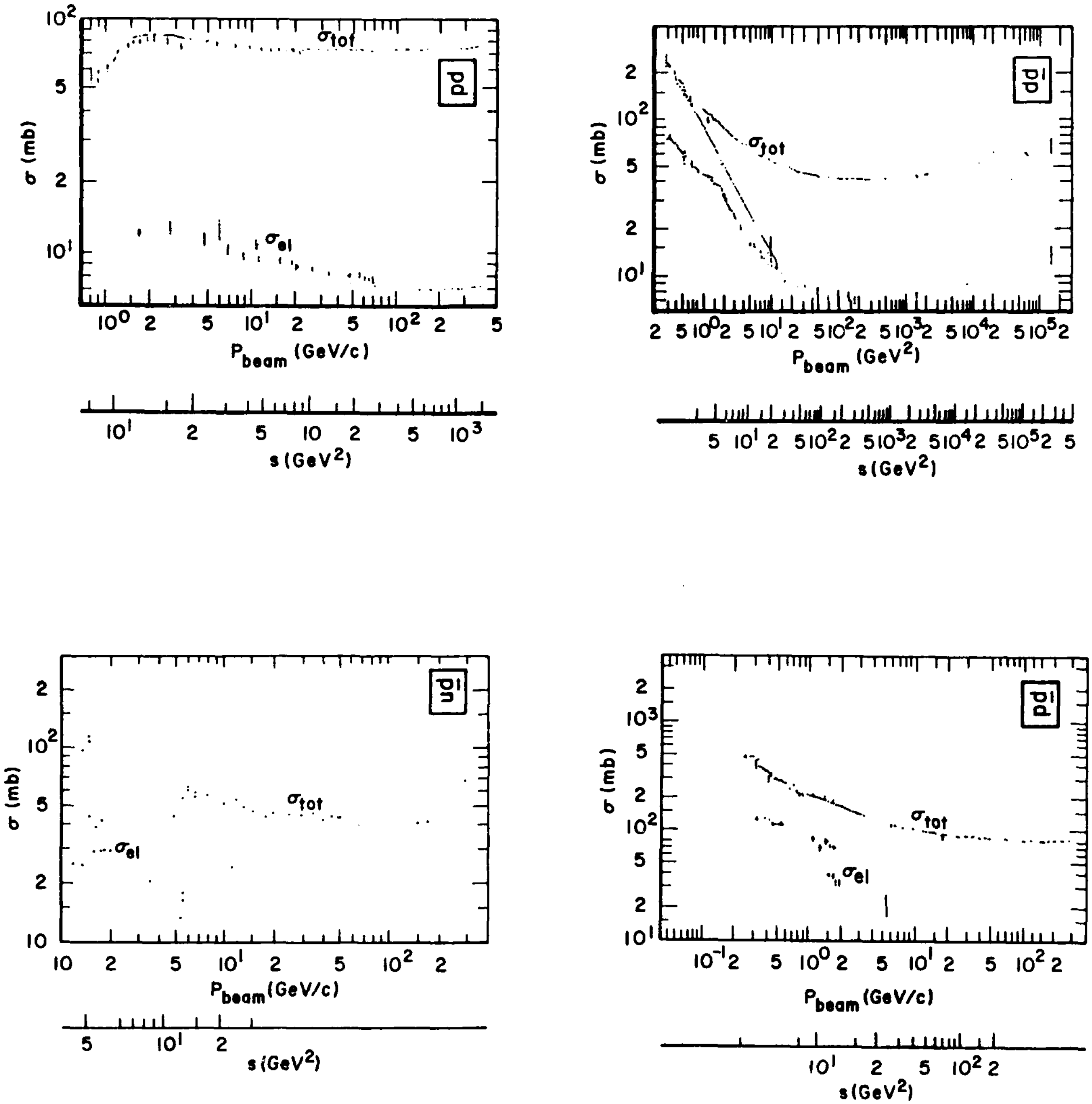


Figure 8. <u>Annihilation Cross-Section Data.</u> (Figure courtesy of V. Flaminio, W. G. Moorhead, D.R.O. Morrison, and N. Rivoure, CERN).

energies decrease rapidly (with increasing energy), they may be disregarded.

The fate of the particle produced by reaction 3, H_{n-1}^{0}, is now considered in the low-temperature, high-magnetic-field environment of the cylindrical "Penning" trap. The resulting cold neutral cluster, H_{n-1}^{0}, may be either even or odd, and have different ortho to para (H_2) ratio. The possible categories are summarized:

Case	Even	Odd	Para	Ortho
1	x		x	
2	x			x
3		x	x	
4		x		x

In cases 1 and 2, the cluster has a single unpaired electron, giving it an appreciable magnetic moment (1 Bohr magneton). The neutral cluster would behave like a very large hydrogen atom and hence all the magnetic schemes developed for H atoms would also apply. Although the cluster would be confined by a shallow axial barrier (near the ends of the magnet), it would drift radially until it encountered something. It is interesting that, if by chance it encountered the pseudo-wall before reaching the ion trap wall, it should be recaptured by the pseudo-wall.

In cases 3 and 4, the cluster magnetic properties are three orders of magnitude weaker than in cases 1 and 2, being due only to the coupling of the nuclear spins (1 nuclear magneton). In case 3, the cluster is composed mainly of parahydrogen; it is diamagnetic (negative susceptibility), has only even rotational levels, and is a "low field" seeker. The magnetic field is essentially constant in the radial direction so the cluster will tend to be gently expelled axially out of the trap. Since the trap is essentially solenoidal, there will be no appreciable radial force. Once more, it may either encounter the pseudo-wall first, or it may just go out of the trap. If the cluster leaves the trap, it could conceivably be recaptured following laser photoionization. In case 4, the resulting cluster is mostly Orthohydrogen; it is paramagnetic,

(positive susceptibility), has only odd rotational levels, and is a "high field" seeker. This means that that there will be a gentle restoring force tending to keep the cluster in the high-field region of the trap. If the initial kinetic energy of the cluster is not greater than this magnetic barrier, the cluster would orbit about the trap center until it encountered the pseudo-wall and recondensed, or hit the trap wall.

The destruction of cluster ions by background hydrogenic species has been found to be something of concern, but does not imply insurmountable difficulties in the pseudo-wall concept.

Very Cold Hydrogen Cluster Ions

If cluster ions of any size may be trapped, one might well ask: Under what circumstances is the Larmor radius smaller than the physical radius of the cluster ion? In this situation the internal motion of the charge on the cluster ion is coupled to the (now very low frequency) cyclotron motion. The remaining degrees of freedom are just a slow $\bar{E} \times \bar{B}$ drift plus axial motion. The whole plasma should be quite dense and fulfill some of the main requirements of a pseudo-wall.

The radius of a spherical cluster ion is

$$r_{ci} = [(3\ V)/(4\ \Pi)]^{1/3} = [(3\ D\ \alpha)/(4\ \Pi)]^{1/3} ,$$

where D is 5×10^{-23} cm^3/molecule and α is the number of molecules in the cluster. The constraint is that r_{ci} be less than the Larmor radius ($r_L = (2\alpha mv)/(qB)$), or

$$r_L < r_{ci},$$

where (2α) is the cluster ion size, m is the mass of a hydrogen dimer, v is the perpendicular component of the cluster ion velocity (includes radial and azimuthal motion), q is the charge, and B is the magnetic field.

This leads to the following inequality;

$$(2\alpha mv)/(qB) < 2.3 \times 10^{-6} (2\alpha)^{1/3} \text{ (meters)}.$$

Putting in values for m, q, and B (8 Tesla), gives

$$[(3.3 \times 10^{-27})/(2.3 \times 10^{-6} \ 1.6 \times 10^{-19} \ 8)] \quad (2\alpha \ n^{2/3} \ v) < 1.$$

Simplification then yields

$$(2\alpha)^{2/3} \ v < 890 \quad \text{(m/s)},$$

where v is in (m/s). This result (which is increasingly less accurate for small α), is given in Table 3 below (with a column of temperatures, assuming a thermal source).

Since the proposed trap will have a liquid helium baffle for cooling, it should be capable of producing this cold, slowly rotating pseudo-wall for cluster sizes α<5000 dimers (or cluster sizes, $2\alpha<10^4$).

TABLE 3. Velocities and Temperatures of Cluster Ions with $r_L < r_{ci}$.

Log(n) (dimers)	v (m/s)	T (K)
1	192.	34.6
2	41.4	16.0
3	8.92	7.46
4	1.92	3.46
5	0.414	1.61
6	0.0892	0.750
7	0.0192	0.346
8	0.00414	0.160

EXPERIMENTAL DESIGN

The system described is similar to the one proposed recently by Kluge et al (Ref. 37). However, since it will contain an annular plasma, it has many of the design features developed by Wong and Rosenthal (Ref. 38).

A generalized schematic of the system is shown in Fig. 9. Note that the main components of the vacuum chamber are in an "L" configuration. It is divided (top to bottom) into the diagnostics, ion trap, 6-way cross, and source chain. To the sides of the 6-way cross are attached the Combo Vac, Cryo Pump (optional), and Star Flange.

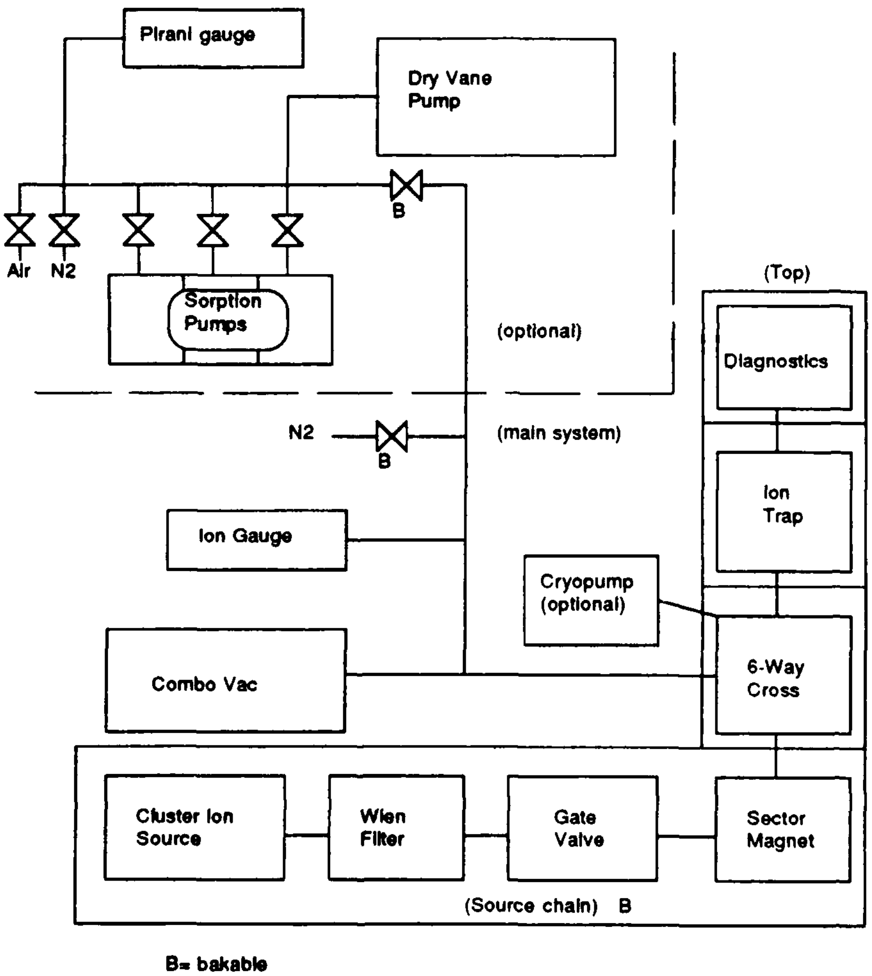


Figure 9. Diagram of the Overall System Layout. The main system is divided into main sections; the source chain constitutes the entire lower section beneath the 6-way cross.

Attached to the bottom is a long, narrow, straight section that makes up the source chain. Note that many of the system components must be bakable (B) to 700 K. The remaining components are the ion gauge and N_2 source. The system is designed to operate satisfactorily with just these mentioned components; however, the optional system, using a mechanical/sorption pump combination is recommended as a later "add on." The addition of the optional system is recommended and would lengthen the times between servicing the Combo and Cryo pumps.

Vacuum Chamber Components

The main components of the vacuum chamber are shown in Fig. 10.

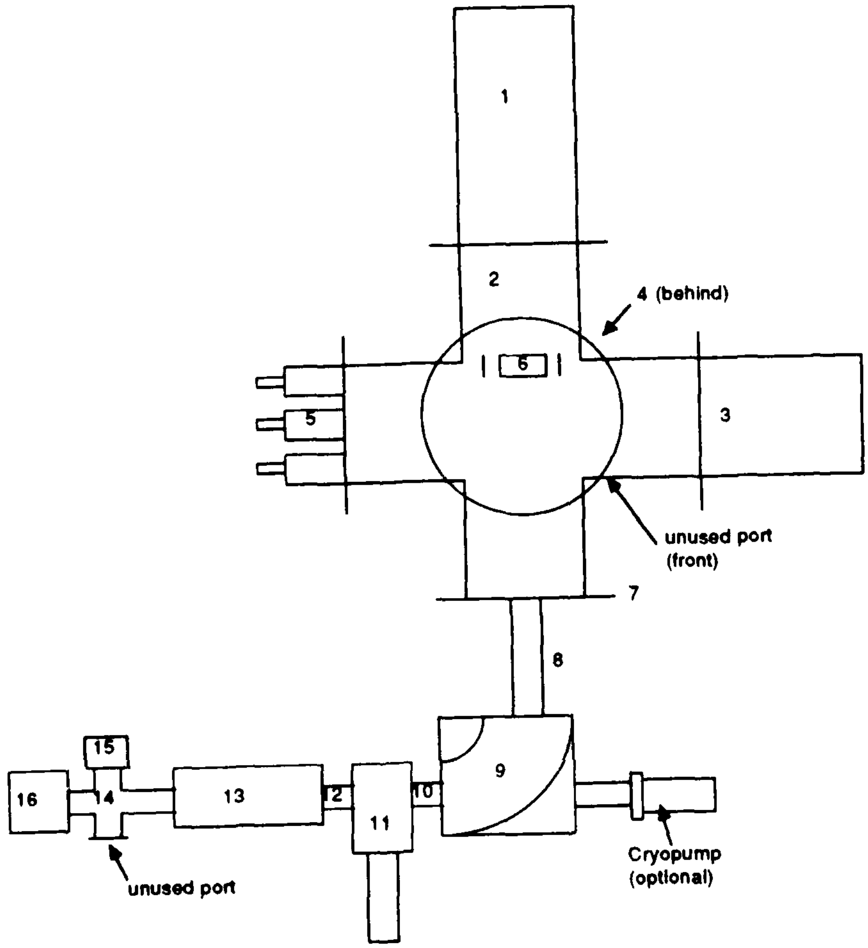


Figure 10. Schematic of the Main System Components. (See text.)

There are 16 essential parts. Unless otherwise noted, 304 stainless steel is to be used for these components, most of which are bakable (excluding pumps) (see Fig. 9). The parts are discussed separately with regard to function and size. This information is from known commercial sources; part numbers were taken from Thermionics Vacuum Products, Hayward, CA; Norcal is suggested as a competitor.

1. The trap wall section is custom built (Fig. 10). It is bakable and features a slip joint lower end (item 7) to allow assembly through the bore of the superconducting magnet (that is, the lower rotatable flange is removable, split into two halves, so that the trap wall section can pass through the magnet bore). This feature will allow easy disassembly. The top flange is a standard rotatable 8 in. conflat.

2. Standard 6-way cross. This component joins the ion trap, combo vac, star flange, optional cryopump, and source chain. One port is unused for access (or future addition).

3. Combo-Vac pump. Commercially available, it comes equipped with a hydrogen diode pump, an 8 in. OD flange, and should be rated at either 750 L/s or 1000 L/s.

3a. 10 in. to 8 in. adaptors will be needed if either the 1000 L/s Combo pump or Cryo-Torr 10 pumps are used.

3b. Bakeout heaters (4) for the 6-way cross. These include controllers.

3c. Liquid nitrogen level controller.

4. Cryopump. The optional cryopump may be added to the available port (suggest CTI Cryo-Torr 10). Space is reserved for this pump, should better vacuum or more gas load (than expected) result.

5. Star Flange. This is commercially available and may be custom built. It provides four smaller ports that will be used for liquid

helium feedthrough (for the liquid helium baffle), controls for the ion trap, N_2 feedthroughs for flooding the system with 1.0 atmospheres of nitrogen, and control of the quadrupole deflector (item 5). This is a 10 in. conflat to 2 3/4 in. (x4) conflat.

5a. One blank 2 3/4 in. conflat.

5b. Nude ionization gauge for monitoring trap pressure. This should have Iridium filaments.

5c Flange for mounting the liquid Helium feeds for liquid helium baffle coolant.

5d. Bakable valve with 2 3/4 in. conflat. Flange with bakable air valve connects to N_2 with safety valve for air vent.

6. Quadrupole lens. This consists of four plates in quadrupolar arrangement and may be home built. Passing the cluster ion beam through this lens causes the beam to take on a helical trajectory. This is used for injecting an annular plasma into the trap.

7. Conflat flange. 10 in. to 4 1/2 in. adaptor.

8. Nipple with 4 1/2 in. conflat.

9. Tunable, Electromagnetic Sector Magnet. This should be custom built by a vendor (suggest Wang NMR) (see also the section on sector magnets). The maximum magnetic field is 0.5 T.

10. Nipple with 4 1/2 in. conflat.

11. Air driven, bakable gate valve with 4 1/2 in. conflat.

12. Nipple with 4 1/2 in. conflat.

13. Tunable Wien filter with electromagnet. This features a yoke for confinement of its magnetic field and should be custom made (suggest Wang NMR) (see also the section on Wien filters).

14. 4-Way Cross with 4 1/2 in. conflat flanges.

15. Nude ionization gauge for monitoring source pressure. This should have Iridium filaments.

16. Hydrogen cluster ion source. An appropriate source that will work is not commercially available and will have to be developed (see section on cluster ion sources).

Table 4 lists the specifications and prices for many of the items.

Fig. 11 shows the locations of the slit assemblies for the Wien filter and the sector magnet. Also given are the specifications for the magnets and electrodes of the Wien filter. Notice that an empty flange is attached to the sector magnet; this is reserved for the addition of a cryopump should the source produce excessive gas load on the pump(s) attached to the 6-way cross.

Also shown is the bakable gate valve used for isolating the trap environment from the source for long-term studies of trapped cluster ions in ultra-high vacuum and the sector magnet.

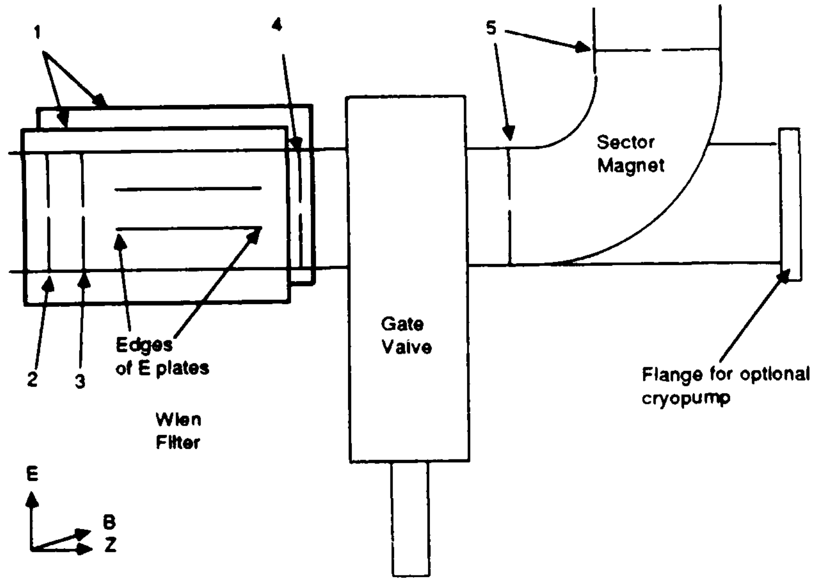
Ion Trap Specifications

The cylindrical ion trap is a modified version of devices used at UCLA and at UCSD. It is designed specifically for large mass-to-charge ratio cluster ions and features a central conducting rod (Fig. 12) that will be used for the neutralization of space charge in the confinement of high-density plasmas. The copper conducting rod (0.6 cm diameter) is supported only at the top, and passes through the end cap grids (end termination grids are used to provide stability for the annular plasma);

TABLE 4. Specifications and Costs of Parts.

Item	Size/Part Number	Unit Price
1	OD 8 in., length 16 in.	$ 900
2	OD 8 in.	3150
3	T#COV750, 750 L/s	8300
	T#COV1000, 1000 L/s	9500
3a	T#1000x800	270
3b	T#IH-100	75
3c	T#LNC-400	475
4		
5	10 in. conflat to four 2 3/4 in.	
5a	2 3/4 in.	14
5b	T#NG-1001	270
5c	Custom	
5d	T#BVV-152	795
5e	T#F275000V	230
6	Custom	
7	T#1000x450	245
8	4 1/2 in.	185
9	Custom	
10	4 1/2 in.	185
11	4 1/2 in. (custom)	1400
12	4 1/2 in.	185
13	Custom	
14	4 1/2 in.	350
15	T#NG-1001	270
16	Custom	

Important note: <u>Due to the the welding and construction techniques used in Ultra High Vacuum systems, this system should never be subjected to more than 1 atmosphere of pressure; doing so could cause leaking.</u>



1. Magnets: Length=20 cm, width=4 cm, spacing=2 cm, B=0.4 T
   Electrodes: Length=20 cm, width=1.2 cm, spacing=5 cm
2. First entrance slit: 5.5 cm from the input electrode edge (1.0mm x 6.0mm)
3. Second entrance slit: 1 cm from input electrode edge (0.3 cm x 6.0 cm)
4. Exit slit: 1 cm from output electrode edge (0.3 cm x 6.0 mm)
5. Sector magnet slits: (1.0mm x 6.0mm)

Figure 11. Schematic Showing a Close View of the Source Chain. Also given are the dimensions of the Wien filter and the five slit assemblies.

it is not electrically connected to the end caps. Notice also that the rod support is hollow so that ions may drift, unencumbered, through the entire trap. Surrounding the central conductor (Fig. 13) is the annular plasma, followed by the inside wall of the trap which is 4 in. in diameter.

The trap electrodes are composed of polished, gold-plated copper (oxygen free) (thickness, 0.7 cm) and are electrically insulated yet thermally connected, via ceramic spacers) to a liquid helium baffle which

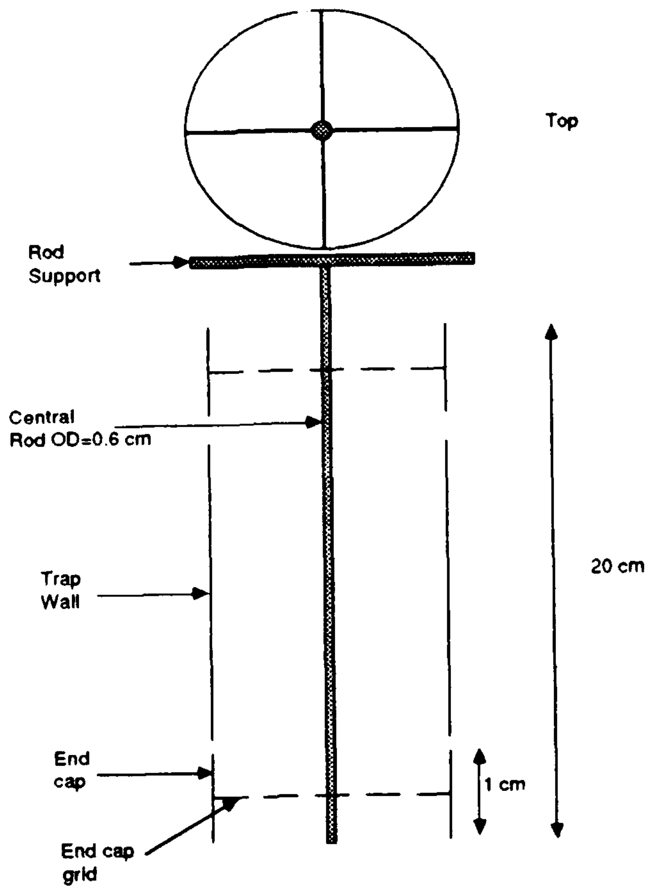


Figure 12. Diagram of the Ion Trap Showing the Trap Walls, End Caps, Grids, and Central Rod.

surrounds it. This was done to eliminate the effects of black-body radiation on stored cluster ions. There is a 1/2 in. gap (1/2 inch in radius) between the trap OD and the inside wall of the liquid helium baffle. The 1/2 in. gap is to prevent heat load on the liquid helium baffle from external sources. This space can also be used for connectors to the diagnostics. The region surrounding the baffle can be filled with porous carbon to improve system pumping of He. The liquid helium baffle is supported by ceramic spacers to the chamber wall (8 in. OD 304 stainless steel). The ID (bore) of the superconducting magnet is roughly 9 in.

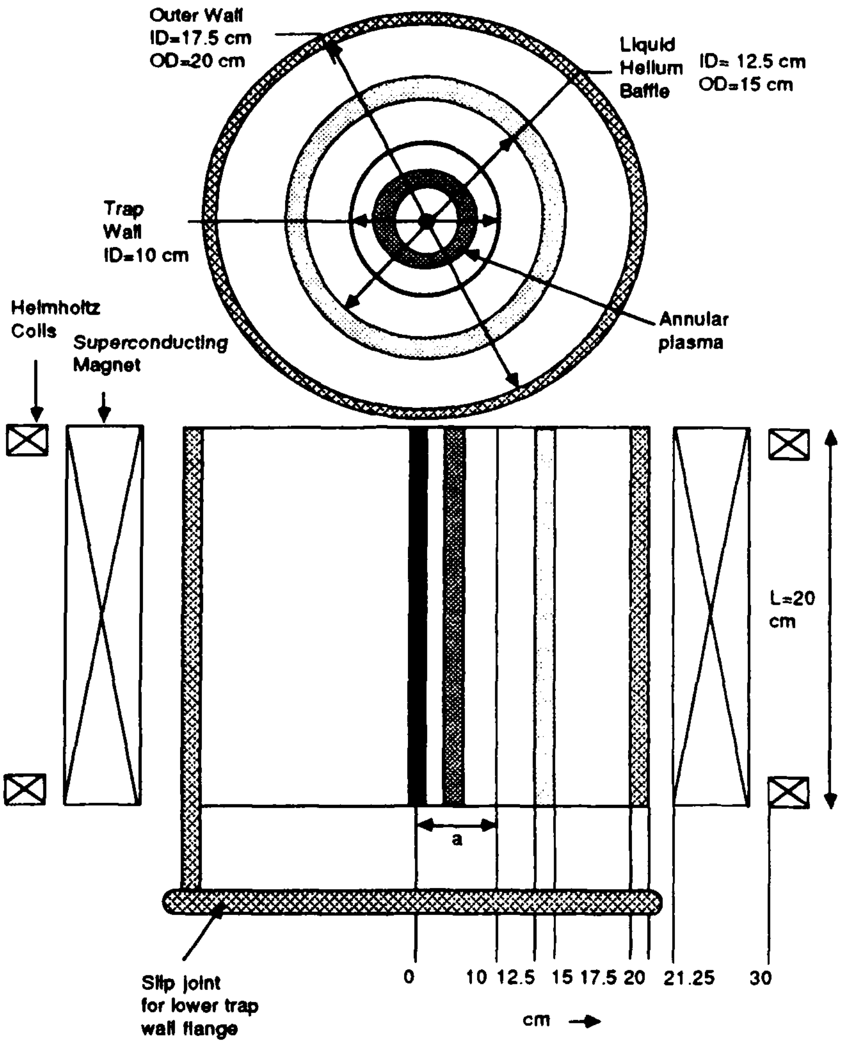


Figure 13. Diagram of the Entire Trap Section Showing the End and Side Projections.

(length 20 cm), allowing room for the slip joint assembly of the lower flange during disassembly of the ion trap wall section. The Superconducting magnet should be custom built to specifications (suggest Oxford). The proposed magnet will use Niobium Titanium for generating fields up to 8.5 Tesla, with long-term use at 7.0 Tesla and with a stability of 3 parts per billion per hour drift. The magnet requires liquid nitrogen and liquid

helium as cryogens. The liquid nitrogen typically requires refill every 7 days (<200 cc/hr), and the liquid helium every 3-6 months (<15 cc/hr). At the outermost diameter are the Helmholtz coils, which may be fabricated at AFAL, and are used to make the magnetic field parallel with the trap axis.

Pumping System

The system pumping requirements were derived through the model shown in Fig. 14a (Ref. 39). P_s (10^{-7} Pa) is the assumed pressure at the cluster ion source, and C_1 is the conductance of the source chain. This is deliberately made very small with the use of small apertures and long length. P_t is the pressure in the trap (10^{-9} Pa). C_2 is the conductance of the channel connecting the trap to the source pump; this is made very large by using a large aperture and short length. P_p and S_p are the pressure and pumping speed of the source pump. The derived relation for S_p is

$$S_p = [(R_w A_w + C_1 P_s) - C_1 P_t]/P_t,$$

where the wall outgassing rate R_w, for baked stainless steel, is 10^{-13} W/m^2, and the inside area A_w of the system is 1.26×10^4 cm^2. Putting in the calculated values for C_1, P_t, and P_s, gives S_p = 5340 L/s (note that this pumping speed is exceeded in the pump specifications). This was done to ensure that the system will be easily able to achieve and maintain pressures in the trap below 10^{-9} Pa. The derived relation for calculating the pressure in the trap is

$$P_t = (C_2 + S_p)(R_w A_w + C_1 P_s)/[(C_1 C_2 + S_p (C_1 + C_2)].$$

The derivation and details may be found in the Appendix.

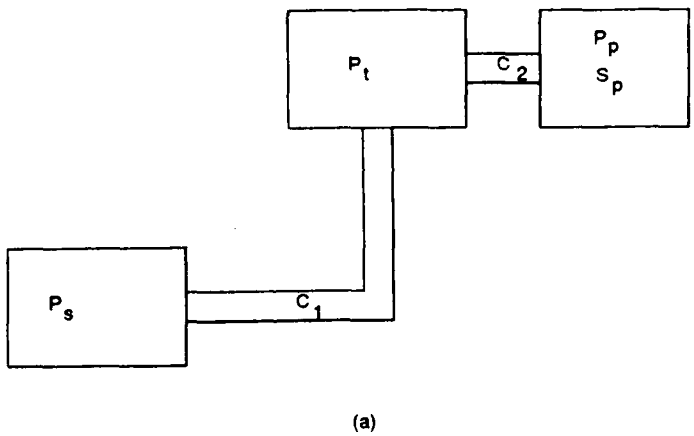
Transit Spreading of the Cluster Ion Beam

In transit from the cluster ion source to the ion trap, the cluster ion beam will undergo a transverse spreading due to space charge

repulsion (Fig. 14b). The maximum allowable transit time of the cluster ion beam pulse is

$$t = 1.52 \times 10^{-3} \ [(R_f - R_i)/R_i] \quad (1/z) \quad (2\alpha/n)^{1/2},$$

where the maximum final radius is R_f (aperture of the ion trap), and the initial beam radius is R_i (aperture of the source), the charge per cluster is z, 2α is the mass number of the cluster (mass of the cluster per proton mass), and *n* is the density of the cluster ions at the source. The constant has units of sec $cm^{3/2}$ so that the answer is in seconds. With this relation, and knowing the distance to the trap, the mass of the transmitted ions, and the pressure of the source, one can easily calculate the accelera acceleration potential ($\Delta V'$) needed for the prescribed transit time. The derivation and further details are given in the Appendix.



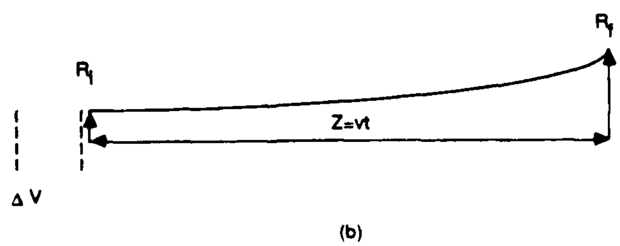


Figure 14. <u>Pumping System and Beam Spread Model.</u> (a) Schematic of the overall system used for the pumping system calculation; (b) Schematic of the model used in the beam spread calculation (See Appendix).

The Wien Filter

The Wien filter is a velocity filter that may use either a fixed or variable magnetic field (the latter field is preferred as an added degree of tuning flexibility). A schematic of a fixed field version[4] of the Wien filter is shown in Fig. 15. This figure is mainly to show device configuration and construction. The dimensions are given in the section entitled "Vacuum Chamber Components." Note that an iron field yoke is used to keep the magnetic field well confined so as to not perturb other system components. Also, not shown, it may be necessary to shield the Wien filter (and sector magnet) from the superconducting magnet by wrapping it with several layers of mu metal. Tn the construction of the Wien filter, a minimum number of feedthroughs and ceramic standoff insulators are used,

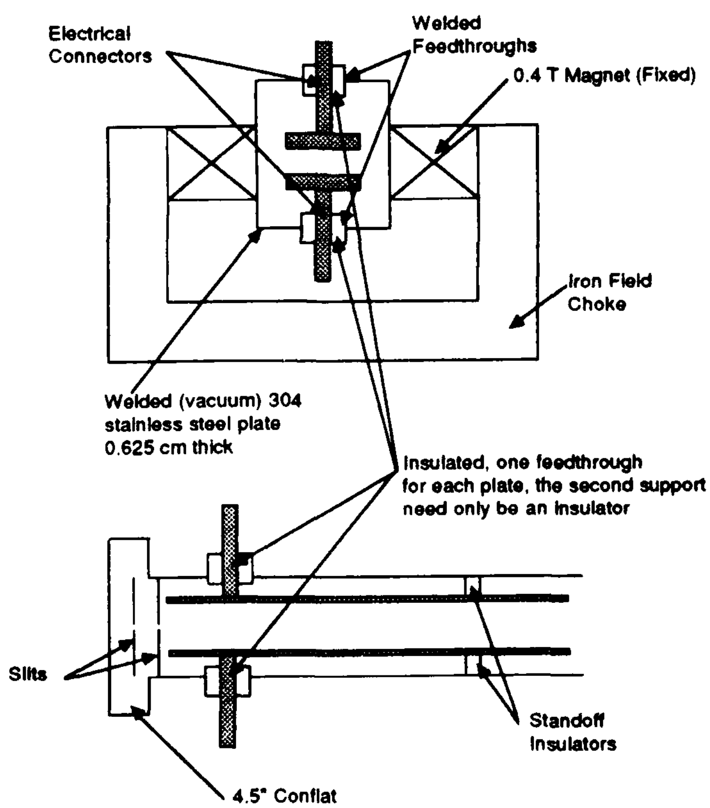


Figure 15. Diagram of the Wien Filter Showing Configuration and Construction Design.

since these have much larger outgassing rates than stainless steel (even when baked). The velocity of the ions transmitted through the filter is given by

$$v = 10^8 V_d'/zB$$

where V_d' is the potential between the electrodes with spacing z, and B is the magnetic field. The mass resolution of the Wien filter is given by (Ref. 40)

$$(\Delta m)/m = (2 z V' w)/ V_d' L^2)$$

where V′ is the potential through which the injected ions have fallen, w is the width of the slits, and L is the length of the electrodes. A mass resolution of 0.01 is deemed sufficient.

The Sector Magnet

The electromagnetic sector magnet is a momentum filter which, when used in combination with the Wien filter, provides an output cluster beam that is both mass and velocity analyzed. A schematic of the device to be built is shown in Fig. 16. Shown are the input, output flanges (and the unused optional flange for the addition of a cryopump), the locations of the slits (dimensions given in the section entitled "Vacuum Chamber Components"), and approximate dimensions. The dashed lines show where the device may be sealed off to exclude unused volume. Again, as with the Wien filter, the device will likely require shielding with mu metal (Ref. 41, 42). The radius of curvature of ions entering the device is given by

$$R = (144/B\beta) (mE)^{1/2}$$

where E is the energy of the entering ions, B is the magnetic field, m is the ion mass, and β is the charge state (+,-). The resolution[40] is given by

$$(\Delta m/m) = w/[R [1-\cos(a)] + x \sin (a)]$$

where, w is the slit width, R is the radius of curvature, and x and a are defined in Fig. 16. Once again, a resolution of 0.01 is considered adequate.

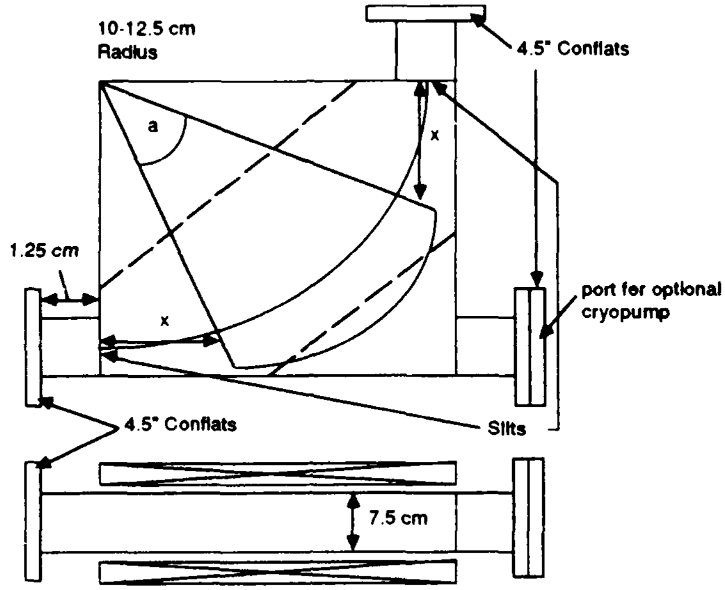


Figure 16. Diagram of the Sector Magnet Showing Configuration and Construction Design.

Since the field produced by the superconducting magnet will be quite strong, a calculation was performed to yield an expression for its variation with distance on axis (Ref. 42). It is necessary to determine how far the source chain, and other devices (such as detectors) must be located to be safe from significant perturbations. The expression for the axial magnetic field is

$$B_z = [(B_o (a^2 + \frac{L^2}{4})^{1/2}) /L] \{ (z + L/2)/((a^2 + (z + L/2)^2)^{1/2}$$

$$- (z - L/2)/((a^2 + (z + L/2)^2)^{1/2}\},$$

where B_o is the field at the center of the magnet, a is the radius of the magnetic coils, L is the length of the magnet, and z is the axial distance from the center of the magnet. Plots of B_z/B_o are shown (linear, followed exponential in B) in Figs. 17 and 18. Notice that for a=20 cm and L=20 cm,

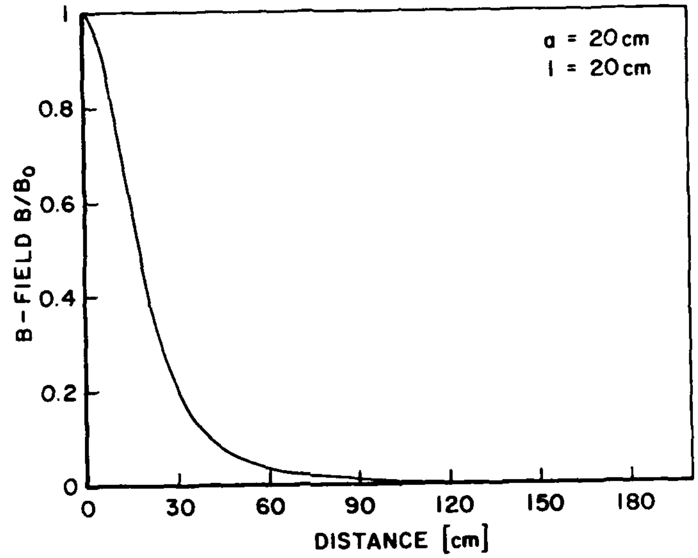


Figure 17. Linear Plot of Magnetic Field Variation Along the Axis of the Superconducting Magnet (Normalized to the Central Field.)

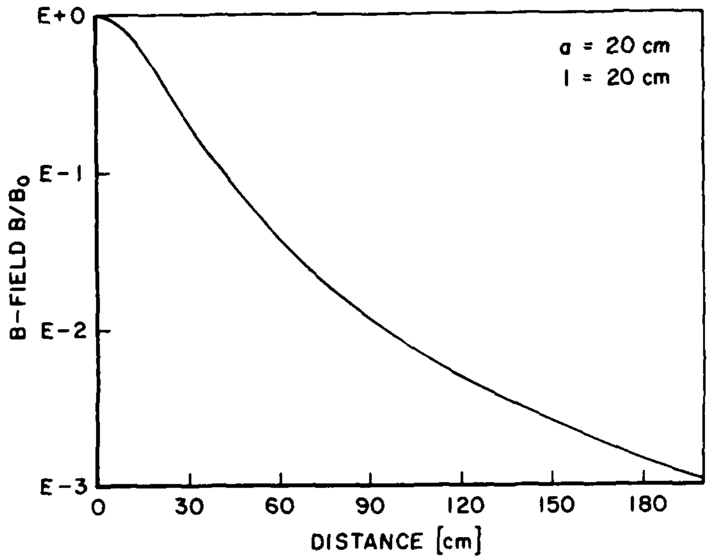


Figure 18. Log Plot of Magnetic Field Variation Along the Axis of the Superconducting Magnet (Normalized to the Central Field).

the axial magnetic field is down by only three orders of magnitude at z=2 meters (still ~ .08 tesla).

Cluster Ion Detection

The detection system chosen depends upon the experiment. Since the initial experiments will measure radial density profiles and total charge, a Faraday cup with charge integrator will be used. The methodology for these measurements is well established in experiments done with pure electron plasmas (at UCSD) and with Lithium ion plasma experiments (at UCLA). The detector will be simply mounted on a standard 4 1/2 in. conflat flange and bolted to the top of the ion trap (Fig. 19).

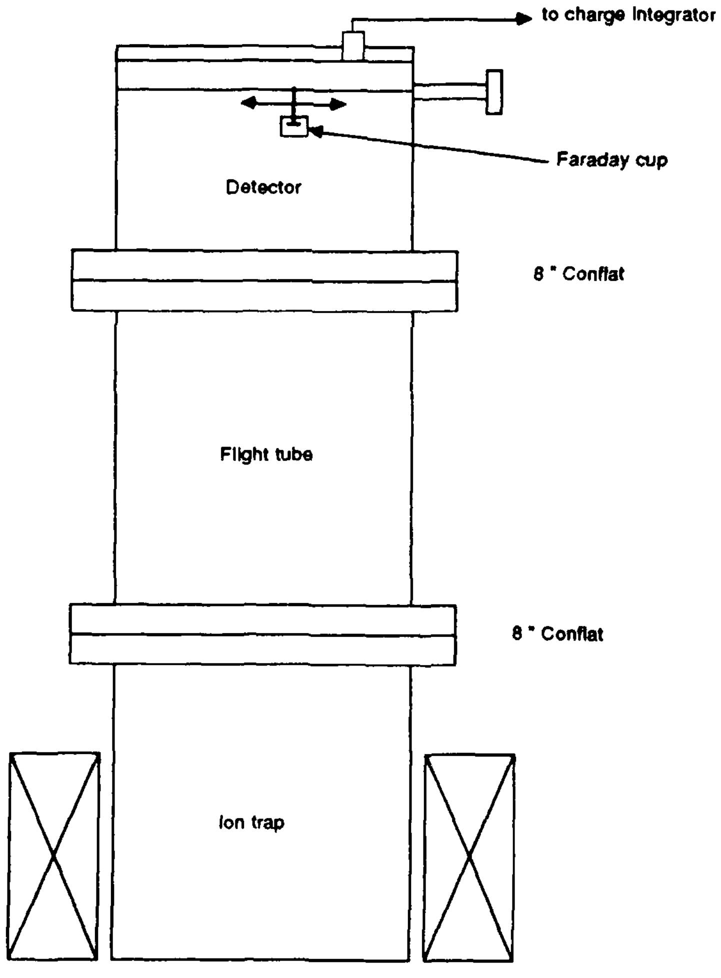


Figure 19. Schematic of the Detection System Intended for Use in the First Experiments.

In this case the output of a radially movable Faraday cup is sent to a charge integrator. Calibration of the detector can be conveniently achieved by running the entire system in "free pass" mode, while using a known source flux. Another useful early experiment is to measure the total stored charge versus time as a function of cluster number. The Faraday cup will also suffice for these measurements.

Ultimately, wishing to determine the thresholds for the A^+ and M^+ growth processes (Refs. 14, 29), one must be able to discriminate the masses of the cluster ions. The steps of a typical experimental scenario would be these:

1. Load the trap with a known mass and number of cluster ions;

2. Add a known background density of reactant, H_2 with known temperature for some time t;

3. Analyze the products for total mass, and charge.

A direct approach to detection uses a setup like the one shown in Fig. 19. At the end of a drift tube is mounted an optional optics stage (not shown), followed by a sector magnet and channel plate detector. The product ions, ejected from the trap with well defined energy, are mass analyzed and detected with a channel plate in combination with a multichannel analyzer. This will give a signal that measures the relative numbers of clusters as a function of cluster mass. Hence, one can observe changes in population of different mass clusters in consecutive single measurements. The total charge can be determined by running the channel plate in total charge mode.

Alternatively, a more elegant method of detection (suggested by A. Y. Wong) uses a narrow band radio frequency source to excite selectively the cyclotron frequency of the ions, driving them to larger Larmor radii until they collide with and are collected on the trap inner surface. One simply attaches a charge integrator to the trap central wall and measures charge as a function of transmitter frequency.

Finally, well known nondestructive detection techniques have been in use for some time in pure electron plasmas (Ref. 43). In one method, the total number of charges can be determined by applying a potential to the end plates. The other method studies ion plasma waves and derives the plasma density.

The Cluster Ion Source

The importance of the cluster ion source to the success of this project should be stressed.

As mentioned, it is not clear that a cluster ion source exists that can fulfill all the requirements. The source should do the following:

1. Provide (a) very large cluster ions, (b) very low cluster temperature, and (c) very high flux;

2. Not use electron impact for ionization;

3. Not load the system with background gas;

4. Operate well around potentially large, stray magnetic fields.

The reasons behind requirements 1 (a) and (b) have been discussed previously. The stringency of requirement 1(b) can be estimated. The current, I, needed to fill the trap is given by

$$I = e\,J\,A = e\,n\,v\,A,$$

where J is the flux, e is the electron charge, n is the density of cluster ions in the trap, v is the velocity of the injected cluster ion beam, and A is the cross sectional area of the cluster ion plasma in the trap. For typical values (in cgs), one obtains

$$I = 10^{-19} \quad 10^{7} \quad 10^{8} \quad 10^{1} = 10^{-3} \text{ amps.}$$

If the total number of clusters arriving at the trap is only 1/10000 of the clusters generated by the source chain (recall Δ m/m = 0.01), then the instantaneous current needed at the source is 10 amps. Calculations indicate that the source need only be on for about 10 microseconds so the minimum estimated total output of the source is roughly 10^{14} molecules of hydrogen.

Three concepts have emerged that could provide an adequate cluster ion source. To our knowledge, none of these sources, as described using hydrogen, have been reported in the literature. In Fig. 20(a), a standard pulsed gas valve is used to generate cluster ions that are simultaneously ionized by a suitable laser pulse. The only thing new here is the use of a laser to ionize the clusters. Since pulsed gas valves cannot be used at temperatures below which viton O-rings become brittle, high pressures of hydrogen gas must be used to achieve adequate cooling of the expanding gas for large cluster production. This necessitates differential pumping and skimming. The retardation grids are added to modify the residence time of the cluster ions. In Fig. 20(b), a solid surface of hydrogen is laser evaporated and laser ionized to produce an intense burst of clusters and ions. This method has been reported to generate cluster ions of carbon, and it has the advantages of low gas load and simplicity. In Fig. 20(c) a refrigerator is used to maintain a bath of liquid hydrogen. A pulse of hydrogen is introduced directly into the liquid from a pulsed valve that is thermally insulated from the cold bath. Finally, the expansion is ionized by a laser pulse. This source should produce high fluxes of very large cluster ions. The use of an inadequate cluster ion source will not prevent the experimental apparatus from working, but can limit its usefulness in studying CC and thus deserves careful attention.

System Performance

This section presents the results of calculations of maximum expected system performance (and plasma parameters) as a function of the cluster number (number of protons in the cluster ion) for the trap. The results presented here are intended to help clarify system capabilities in terms of

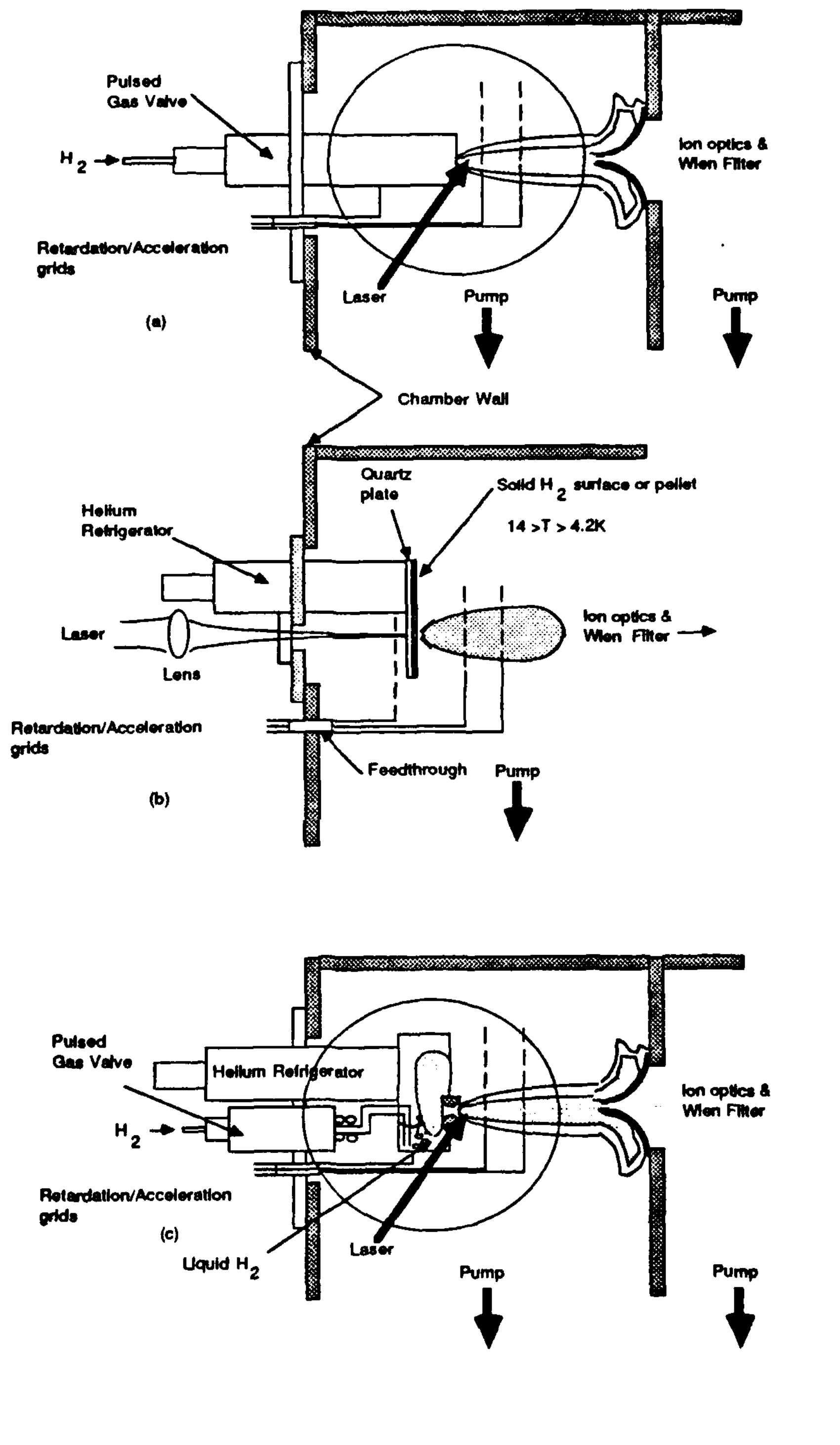


Figure 20. <u>Proposed Cluster Ion Sources.</u> (a) Pulsed gas valve/laser cluster ion source; (b) Solid surface/laser cluster ion source; (c) Pulsed gas valve evaporation/laser cluster ion source.

scalability to large sizes. The trap parameters used here are nearly the same as those of the proposed device, with the exception of length (the proposed device is 20 cm long). The parameters used in these calculations are

B: The magnetic field (10 T).

Φ: The potential applied to the central conductor (1000 Kvolts).

a: The radius of the central conductor (0.3 cm).

d: The radius of the trap wall (5 cm).

r: The radius of the plasma (2.5 cm).

Δ: The thickness of the (annular) plasma (0.2 cm).

L: The length of the magnet (100 cm).

Fig. 21 shows the variation of the cyclotron frequency with cluster number. The cyclotron frequency is given by

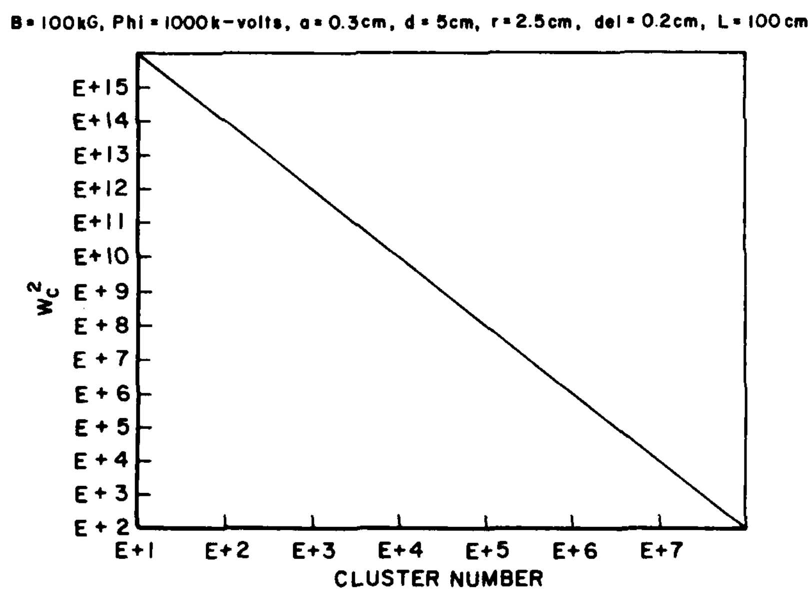


Figure 21. Cyclctron Frequency Versus Cluster Number.

$$W_c = eB/mc,$$

where e is the charge, B is the magnetic field, m is the cluster mass, and c is the speed of light. Notice that it is linear and is only about 100 Hz for clusters of size number 10^7.

Fig. 22 is obtained from the density limit relation (Ref. 44),

$$W_c^{\,2} - (4\ e\ \Phi),(m\ R^{+2}\ \ln(d/r)) = W_p^{\,2}\ [\ \{(8\ R\ \Delta)/R^{+2}\} -$$

$$\{(4\ R\ \Delta - 2\ R^{+2}\ \ln(R^{+}/r) + 2\ R^{-2}\ \ln(R^{-}/r)\}/\{R^{+2}\ \ln(d/r)\}].$$

In Fig. 22 the quantity

$$\mathrm{Phi} = \frac{4\ e\ \phi}{M\ R^{+2}\ \ln\ (d/r)}$$

B = 100kG, Phi = 1000k-volts, a = 0.3cm, d = 5cm, r = 2.5cm, del = 0.2cm, L = 100cm

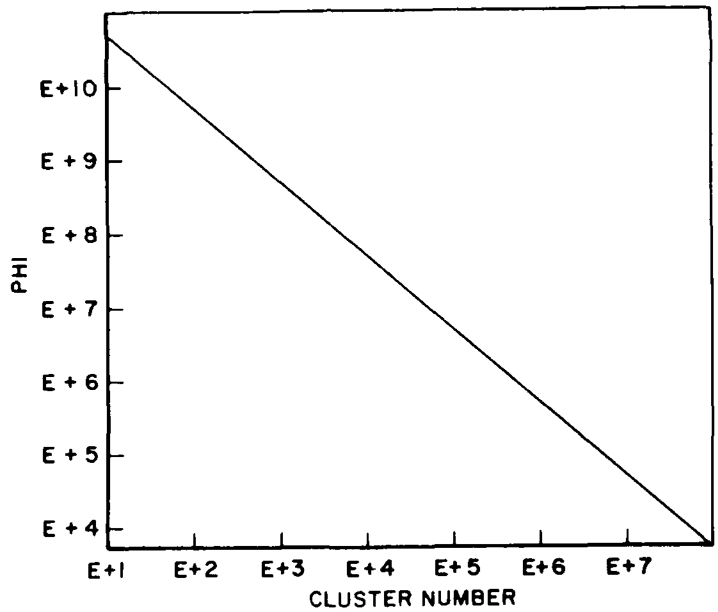


Figure 22. Central Rod Potential Versus Cluster Number at Constant Total Mass.

is plotted versus cluster number. This quantity is indicative of confinement potential of the bias (1000 KV) on the central conduction and has units of sec.$^{-2}$.

Definitions of the variables are the following: W_c is the cyclotron frequency, e is the charge, Φ is the potential on the central rod, m is the mass, R^+ is the outside radius of the plasma, d is the radius of the trap, r is the radius of the plasma (to its center), twice Δ is the width of the plasma, and R^- is the inside radius of the plasma. The linear decrease is just the result of less space charge repulsion per mass as the size of the clusters increases.

Fig. 23 shows the variation in the characteristic plasma frequency with increasing cluster number (at constant trapped mass). The plasma

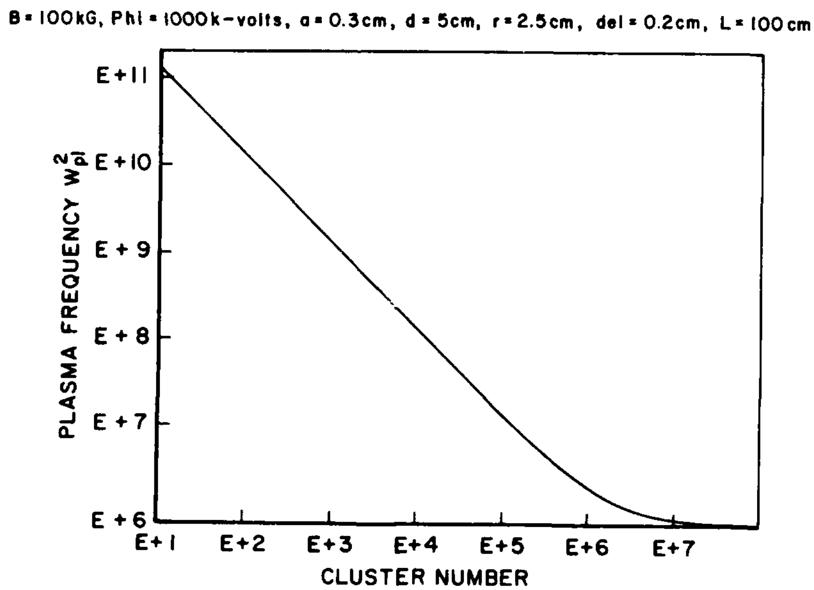


Figure 23. <u>Plasma Frequency Versus Cluster Number.</u>

frequency is given by

$$w_p^2 = (4 \Pi n e^2)/(e_o m),$$

where, Π is 3.14, n is the plasma density, e is the cluster charge, e_o is the permittivity of free space, and m is the cluster mass. Notice that at a cluster size near 10^6 the plasma frequency reaches a minimum plateau. The plateau indicates the increasing dominance of electrostatic confinement for high cluster number. It also indicates a balance between the number of charges on the central conductor and the number of charges in the plasma.

Fig. 24 is a plot of the confinable number density that may be contained in the trap for the given magnetic field and central conductor bias. Note that the density needed begins to remain constant at about 10^6 cm^{-3} for mass numbers above 10^7. Fig. 25 is the same as 24, except given in terms of mass per volume.

Fig. 26 is a plot of the total confinable mass in the trap as a function of the cluster number. Notice that, with large cluster sizes (of around 10^6), the total stored mass increases sharply.

Fig. 27 is the same as 26 except with a much larger trap. The trap parameters in this case are

B: The magnetic field (100 T).
Φ: The potential applied to the central conductor (1000 Kvolts).
a: The radius of the central conductor (50.0 cm).
d: The radius of the trap wall (100 cm).
r: The radius of the plasma (80.0 cm).
Δ: The thickness of the (annular) plasma (6.0 cm).
L: The length of the magnet (200.0 cm).

Notice that with this (only somewhat futuristic) trap, the storage of greater than 0.3 mg of cluster ions is possible in a device only two meters long. This corresponds to an energy density of 5×10^6 J/m^3.

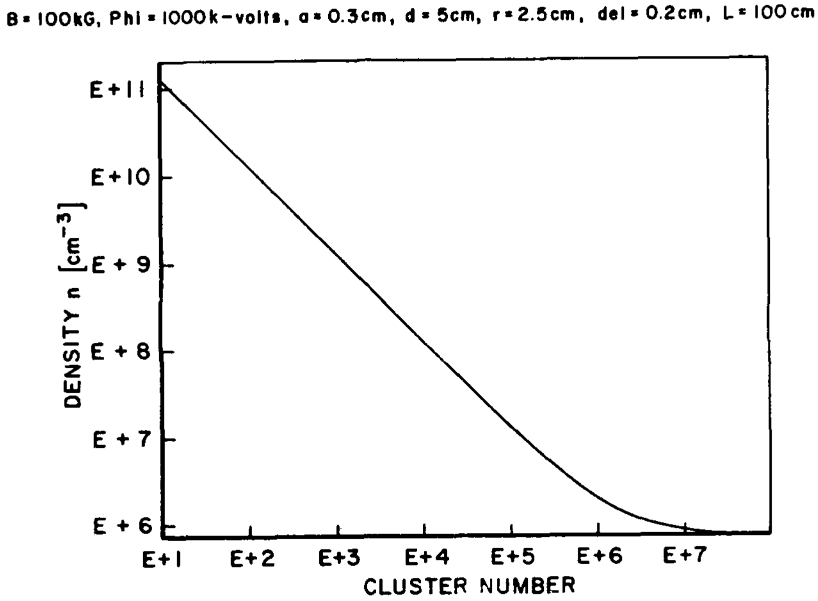


Figure 24. Plasma Number Density Versus Cluster Number.

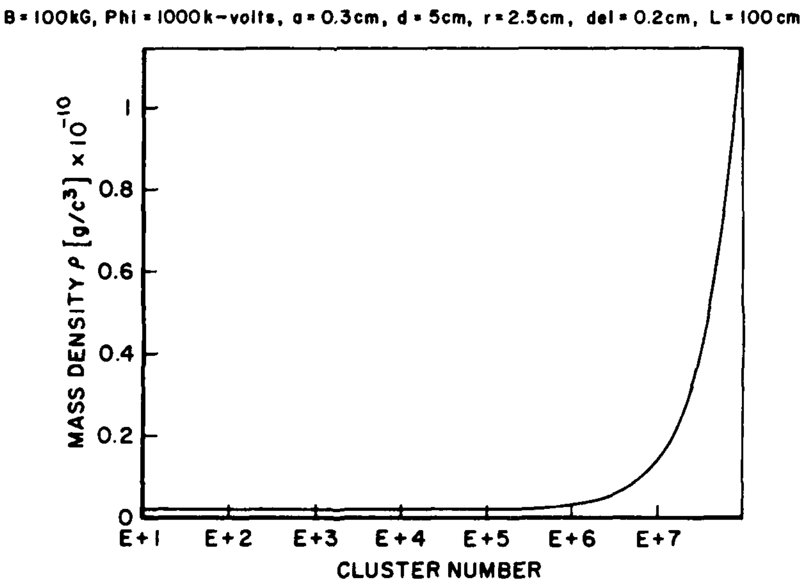


Figure 25. Mass Density Versus Cluster Number.

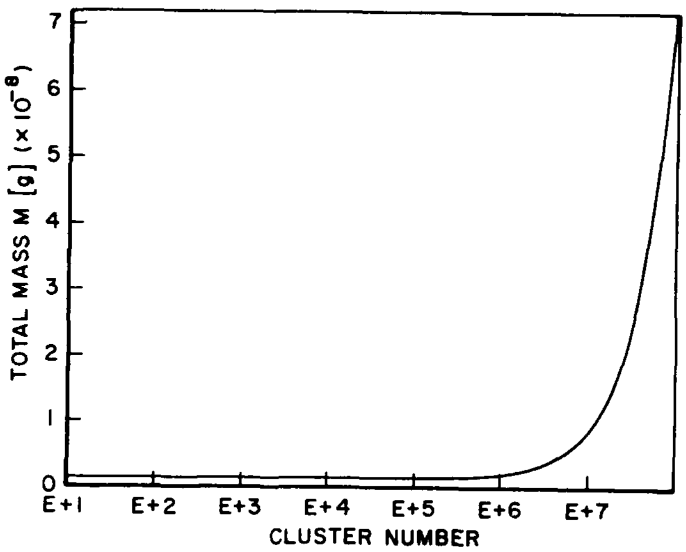


Figure 26. Total Mass Confined by the Trap Versus Cluster Number for a 100-cm Trap. (Note the dramatic increase beyond cluster numbers of 10^7.) This trap is 5 times longer than the one proposed; the remaining parameters are similar.

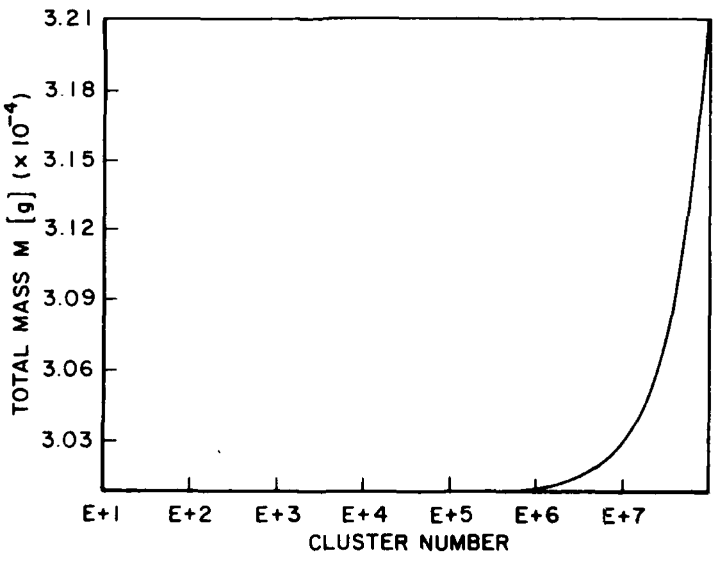


Figure 27. Total Mass Confined by the Trap Versus Cluster Number for a 300-cm "Next-Generation" Trap. (Note the dramatic increase beyond cluster numbers of 10^7.) This trap is 10 times longer and the field is higher than the one proposed; the remaining parameters are similar.

CONCLUSIONS AND RECOMMENDATIONS

In conclusion, the research effort for the past two years has resulted in the development of the CI approach to CC (Containerless Condensation). The general strategy in the years ahead is to develop CC techniques, more specifically, to build the first pseudo-wall designed for long-term studies of very large hydrogen cluster ions. If successful, this will fulfill the near-term goal of this project: Implementation of the experimental investigation and understanding the formation of hydrogen cluster ions from the viewpoint of "containerless" condensation.

Recommendations pertaining to the construction and operation of the cluster ion trap have been made in the previous section. It is anticipated that the program will require one year to construct and test the basic apparatus. The first experiments (previously stated) should start the second year. The remainder of this section gives an approximate timetable covering the next three years.

The effort for the first year will involve the purchase, installation, and checkout of the UHV system. This should include all of the components given in Fig. 10. The Wien filter and sector magnet should also be built by the appropriate subcontractors. One of the two optional cryopumps could be purchased and attached either at the 6-way cross or the sector magnet.

Professor A. Y. Wong and Dr. G. Rosenthal should be consulted on matters concerning system construction. Professor Wong and Dr. Rosenthal have agreed to set aside part of their time this year for this purpose.

Also, during the first year, needed experimentation on the cluster ion source should be carried out. Low background flux, pulsed sources should be employed exclusively. Initially, a commercial source manufactured by Coultron was under consideration, but this was abandoned because of the need for too many modifications and limited mass/charge handling capability.

Some construction on the ion trap should also begin the first year.

The cryopump is the final stage of pumping, used for obtaining very high vacuum at high pumping speed.

The second year should involve the construction and implementation of the cluster ion source and ion trap. The source and most of its components should be constructed and tested on site. The ion trap should be constructed by an appropriate vendor (suggest UCLA machine shop) under the care and supervision of persons with first-hand experience. The surfaces will be machined to high tolerence from a single supply of oxygen-free copper. Special care is required in the mounting and assembly to avoid field inhomogeneities, junction potentials, and stray capacitances. The trap inner sections could also be electroformed. This is more expensive, but yields a better surface.

The superconducting magnet (8.5 Tesla) should be custom made by an experienced commercial vendor (recommend Oxford). They can also provide the liquid helium baffle and all of the supporting equipment for the magnet.

The detection hardware should be purchased and assembled on site during the second year. If all goes well, the system should be ready for checkout near the end of the second year.

With the system near completion, the third year will involve performing the first experiments. As mentioned previously, these will measure the plasma radial density profiles, the total charge in the trap, and the confinement times as a function of cluster size.

In later experiments, the ion trap will be filled with cluster ions of known size and temperature. Reactant (cold H_2 or H) will be added to the cooled cluster ion pseudo-wall. Cooling cycles may be needed during the addition of the reactants. After some time, the ion cloud composition will be analyzed by releasing the trap contents for mass and energy analysis. The cycle is then repeated. Cluster ion growth rates and critical information are then determined from the data.

In the fragmentation experiments, the scenario is similar. The ion trap is again loaded with cluster ions of known composition and temperature. They are then exposed to an appropriate collider (protons, antiprotons, positrons, photons, etc.) of well defined energy. The composition that remains in the trap is then mass and energy analyzed. The results of the fragmentation studies will decide if large cluster ions can be conveniently fragmented into just a few large charged pieces (with minimum H or H_2 loss).

A series of experiments should be done that involve the multiplication of seed crystals by fragmentation. The general fragmentation reaction using protons is

$$\{H_p^+\} + K\,[H^+] = \{K\,[H_{p+1/k}^+]\}.$$

The general fragmentation reaction for photons is

$$\{H_p^+\} + K\,[hv] = \{K\,[H_{p+1/k}^+]\} + k\,e^-,$$

where k and n are integers. Note that the resultant fragments must remain captured and cooled. The first fragmentation reaction has the highest priority.

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APPENDIX A

Radial Confinement and the Classical Brillouin Limit

The presented derivation may be found in R. C. Davidson's book (Ref. 45).

For the confinement, a charged particle undergoing uniform circular motion in a stable orbit that lies in a region within electric and magnetic fields, the centrifugal force must balance the Lorentz force (see Fig. A-1),

$$-\frac{m\,v^2}{r} + e \quad E_r^o + v_\theta^o\,B = 0 \tag{A.1}$$

where E_r^o and v_θ^o are the radial electric field and azimuthal velocity at equilibrium, m is the mass, r is the orbit radius, and B is the magnetic field.

Since the radial electric field is self-generated by the plasma, it must be determined self-consistently using Poisson's equation,

$$\frac{1}{r}\,\frac{\quad}{r}\,r\,E_r^o = -4\,\pi\,e\,n^o(r) \quad . \tag{A.2}$$

The simplest solution is to take $n^o(r)$ to be constant; then the electric field may be obtained through direct integration giving

$$E_r^o = -r\,4\,\pi\,e\,\frac{\bar{n}}{2} \quad , \tag{A.3}$$

for $0<r<R_p$. This may be expressed in terms of the plasma frequency

$$\omega_p^2 = \frac{4\,\pi\,\bar{n}\,e^2}{m} \quad , \tag{A.4}$$

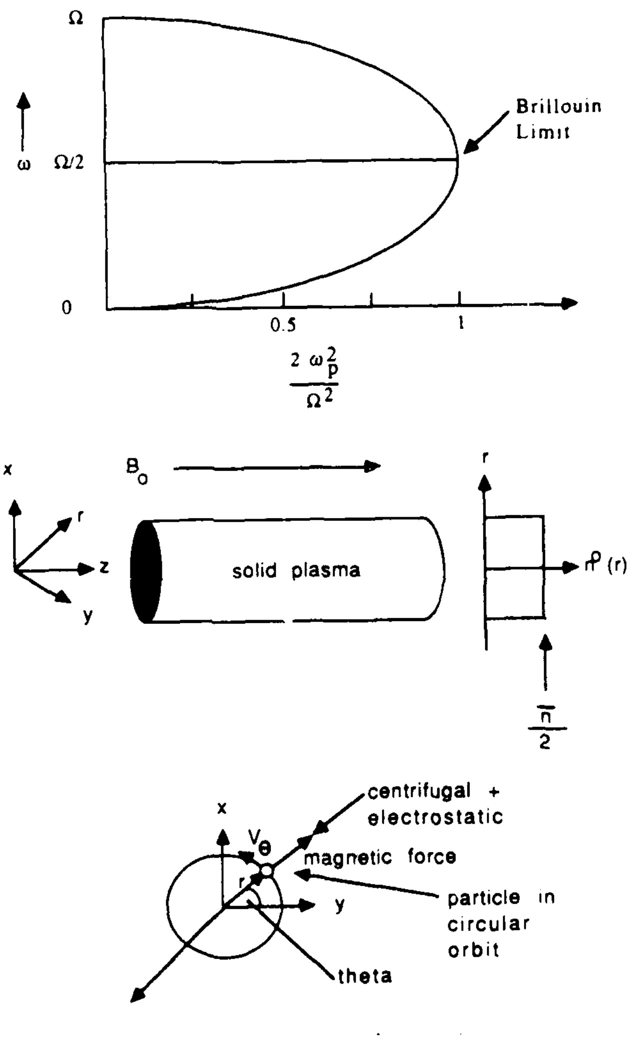

Figure A.1. <u>Plot of the Two Rotational Frequencies of a "Solid" Plasma and the Location of the Brillouin Limit.</u> Below are shown the conventions used in the derivation presented in the text (Appendix).

as

$$E_r^o = \frac{m\,\omega_p^2\,r}{2\,e} \tag{A.5}$$

for $0<r<R_p$. Introducing the angular frequency

$$\omega_v = \frac{v_\theta^o}{r}\ , \tag{A.6}$$

and the cyclotron frequency,

$$\Omega = \frac{e\,B_o}{m\,c}\ , \tag{A.7}$$

into equation (A.1 , it may be expressed as

$$\omega^2 + \Omega\omega + \frac{\omega_p^2}{2} = 0\ . \tag{A.8}$$

The solutions to this quadratic are

$$\omega = \omega^{+-} = \frac{\Omega}{2}\left[1 \pm \left(1 - \frac{2\,\omega_p^2}{\Omega^2}\right)^{\frac{1}{2}}\right]\ . \tag{A.9}$$

The two rotational frequencies ω^+ and ω^- are plotted in the figure as a function of $\frac{2\,\omega_p^2}{\Omega^2}$.

In the low density limit the fast rotation, ω^+, corresponds to all the particles rotating about the axis of symmetry at the cyclotron frequency. The low frequency, ω^-, corresponds to the trochoidal orbits which give rise to a slow $E^o \times B_o\, e_z$ rotation of the plasma.

The high density limit is known as the Brillouin limit. It can be seen from Equation (A.9) that when $\frac{2\,\omega_p^2}{\Omega^2} = 1$, then

$$\omega^+ = \omega^- = \frac{\Omega}{2}\ , \tag{A.10}$$

Furthermore, for the solutions to Equation (A.9) to be real, we must have,

$$\omega_p^2 < \frac{\Omega^2}{2} . \tag{A.11}$$

This equation states that the electrostatic repulsive forces, measured by ω_p^2 must be overcome by the magnetic restoring force, measured by Ω^2. Inserting the definitions for w_p^2 and Ω^2 into Equation (A.11) yields the relation

$$n\, m\, c^2 < \frac{B^2}{8\pi} , \tag{A.12}$$

where n is the density, m is the mass of the cluster ion, and B is the magnetic field. This equation gives the limit to the mass density (nm) that can be achieved under the assumption of a solid cylindrical cylinder with constant radial density. This limit may be exceeded in two ways:

1. By operating in the low density (n), high mass (m) regime (see plots in the section entitled "System Performance").

2. By using an *annular plasma*, neutralized by a central conducting rod.[37,43]

System Pumping

As discussed in the section of the report entitled "The Pumping System" (see Fig. 14a) the pressure in the trap is given by

$$P_t = \frac{P_s}{1 + \frac{C_2}{C_1} \quad \frac{S_p}{S_p + C_2}} , \tag{A.13}$$

where P is the pressure in the source or trap (Pa or Torr), C's are the conductances (L/s), and S the speed of the pump (L/s). We want to include the outgassing rate of the baked stainless steel (10^{-13} W/m^2) in the derived expression for the pumping speed.

Define

$$S_p' = \frac{s_p \; c_2}{s_p + c_2} \;, \tag{A.14}$$

then

$$P_t = \frac{P_s}{1 + \dfrac{s_p'}{c_1}} \;, \tag{A.15}$$

or

$$P_t = \frac{c_1 \; P_s}{c_1 + s_p'} \;. \tag{A.16}$$

Now, P=Q/C, and applying this to the source and trap gives

$$P_s - P_t = \frac{Q_1 + Q_w}{c_1} \;, \tag{A.17}$$

rearranging gives,

$$c_1 \; P_s = c_1 \; P_t + Q_1 + Q_w \;; \tag{A.18}$$

inserting this into equation (A.16) gives

$$P_t = \frac{c_1 \; P_t + Q_1 + Q_w}{c_1 + s_p'} \;; \tag{A.19}$$

solving this expression for P_t gives

$$P_t = \frac{Q_1 + Q_w}{s_p'} \;, \tag{A.20}$$

$$= \frac{c_1 \; (P_s - P_t) + \alpha_w \; A_w}{s_p'} \tag{A.21}$$

$$= \frac{\alpha_w \; A_w + c_1 \; P_s}{s_p' + c_1} \tag{A.22}$$

$$= \frac{\alpha_w A_w + c_1 P_s}{c_1 + \frac{s_p c_2}{c_2 + s_p}} \tag{A.23}$$

$$= \frac{(c_2 + s_p)(\alpha_w A_w + c_1 P_s)}{c_1 (c_2 + s_p) + s_p c_2} \tag{A.24}$$

$$= \frac{(c_2 + s_p)(\alpha_w A_w + c_2 P_s)}{c_1 c_2 + s_p (c_1 + c_2)} \tag{A.25}$$

Now, solving this expression for s_p gives

$$s_p = \frac{(c_2 + s_p)(\alpha_w A_w + c_1 P_s) - c_1 c_2 P_t}{P_t (c_1 + c_2)} \quad . \tag{A.26}$$

The strategy in the system design is to make the conductance of the source chain (c_1) very small relative to c_2 (which is made very big), and this can be achieved to a good approximation in the design presented. Thus, taking the appropriate limit gives

$$S_p \ \mathrm{Lim}_{c_2} = \frac{\alpha_w A_w + c_1 P_s - c_1 P_t}{P_t} \quad , \tag{A.27}$$

Specifying the parameters on the right side as

$$\alpha_w = 10^{-13} \frac{W}{M^2} \tag{A.28}$$

$$A_w = 1.26 \ M^2 \tag{A.29}$$

$$P_s = 10^{-9} \ \mathrm{Torr} \tag{A.30}$$

$$P_t = 10^{-11} \ \mathrm{Torr} \tag{A.31}$$

$$c_1 = 121 \frac{d^3}{L} = .054 \frac{M^3}{s} \quad . \tag{A.32}$$

This finally gives the pumping speed as

$$S_p = \frac{1.26 \times 10^{-10} + 7.18 \times 10^{-6}}{1.33 \times 10^{-9}} - 54. \frac{L}{s} \tag{A.33}$$

$$S_p = 5344 \ . \frac{L}{s} \ . \tag{A.34}$$

Transit Spreading of the Beam

This "worst case scenario" calculation assumes that a cluster ion plasma of density n_o, mass m, charge q is formed instantaneously and then expands in the x direction under the influence of the self electric field, E. The equation of motion is

$$\begin{aligned} m\,x &= q\,E \\ x &= \frac{q\,E}{m} \end{aligned} \tag{A.35}$$

Inserting the electric field, $E = (mR/2q)\ w_p^2$, gives

$$a = x = \frac{q\,m\,R\,4\,\pi\,q^2\,n_o}{m\,2\,q\quad m} = \frac{2\,\pi\,q^2\,n_o\,R}{m} \tag{A.36}$$

$$a = \frac{\omega_p^2\,R}{2}$$

where the plasma frequency is given by

$$\omega_p^2 = \frac{4\,\pi\,q^2\,n_o}{m} \quad . \tag{A.37}$$

The Newtonian equations of motion are

$$\begin{aligned} v &= v_o + a\,t \\ v &= x_o + v_o t + \frac{1}{2}\,a\,t^2 \quad . \end{aligned} \tag{A.38}$$

The initial conditions are

$$v_o = 0 \text{ and } x_o = R_i \quad . \tag{A.39}$$

Where R_i and R_f are the radii of the source and the ion trap respectively,

$$R_f = R_i + \frac{1}{2} a\, t^2 \tag{A.40}$$

$$(R_f - R_i)\ 2 = \frac{\omega_p^2\, R_i}{2\, t^2} \tag{A.41}$$

solving for t gives

$$t^2 = \frac{4\ (R_f - R_i)}{R_i\ \omega_p^2} \tag{A.42}$$

$$t = 2 \left(\frac{R_f - R_i}{R_i}\right)^{\frac{1}{2}} \left(\frac{1}{4\,\pi}\right)^{\frac{1}{2}} \left(\frac{m}{q\ n_o}\right)^{\frac{1}{2}} \tag{A.43}$$

This equation gives the maximum allowable transit time for the beam of cluster ions. It may be recast into a more convenient form, by defining 2α as the mass of a cluster divided by the mass of a proton, z as number of proton charges per cluster, and n_i as the number density of cluster ions (cm^{-3}).

$$t = 1.52 \times 10^{-3} \left(\frac{R_f - R_i}{R_i}\right)^{\frac{1}{2}} \frac{1}{z}\ \frac{2\alpha}{n_i}\ . \tag{A.44}$$

The constant in front has the dimensions ($s\ cm^{3/2}$). Typical values for R_i and R_f, the cluster sizes, and densities give times in the milliseconds. The length the cluster ions will travel is roughly 2 meters, implying a cluster ion injection velocity of 2×10^5 cm s^{-1}. This is easily achieved.

APPENDIX B

Bibliography of Hydrogen Cluster Ions

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Introduction

The enclosed bibliography includes the edited results of September 1987 Chemical Abstracts On-Line Searches, plus selected additional references, for the period 1966-1987 for the species H_2^+ (and H_2^-), H_3^+ (and H_3^-), and H_N^+ and (H_N^-, $N \geq 4$), in Sections I, II, and III, respectively. References are roughly in reverse chronological order. Note that H_2^- is not a stable species and the evidence suggests H_3^- is also unstable. All references have been copied or requested through interlibrary loan. The assistance of Todd Colin and Tom Yang in these searches and partial support from the Rand Corporation are gratefully acknowledged.

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